

Philips Technical Review

DEALING WITH TECHNICAL PROBLEMS
RELATING TO THE PRODUCTS, PROCESSES AND INVESTIGATIONS OF
THE PHILIPS INDUSTRIES

THE "ELECTRONIC POEM" PERFORMED IN THE PHILIPS PAVILION AT THE 1958 BRUSSELS WORLD FAIR

A. THE LIGHT EFFECTS, by L. C. KALFF

B. THE SOUND EFFECTS, by W. TAK

C. THE ELECTRONIC CONTROL SYSTEM, by S. L. de BRUIN.

628.973: 534.86: 621.316.7.076.7: 789.983

In its pavilion at the 1958 Brussels World Fair Philips has broken with tradition, in that its products are not put on display as is customary at other exhibitions and trade fairs, but are symbolized, as it were, by a novel synthesis of light and sound — Le Corbusier's "Poème Electronique". Among those who collaborated with Le Corbusier in achieving this synthesis were the composer Varèse, the film producer Agostini and the technical specialists contributing to this article. The creation of this "Electronic Poem" called for elaborate lighting and electro-acoustical equipment; moreover, since it is performed and repeated almost entirely automatically, an intricate, largely electronic, control system was needed.

A. THE LIGHT EFFECTS

by L. C. KALFF.

Philips is represented at the Brussels World Fair by a pavilion designed by Le Corbusier and Xenakis. The highly unusual architecture of the pavilion is described in a series of articles published in the previous issue of this journal¹⁾. In the present article we shall be concerned with what takes place inside the structure. Here the public is shown how science and technology have created an instrument which provides the artist with a novel form of expression using light and sound effects. Audiences of some

five hundred at a time see a performance of the "Electronic Poem" by Le Corbusier and his collaborators. On the curved walls the audience sees a continuously shifting play of projected pictures and colours. These are accompanied by sound effects which, by means of electro-acoustical tricks, join in creating the illusion of movement.

In this "Electronic Poem", Le Corbusier has attempted to express his view that our increasingly mechanized civilization is striving towards a new harmony. The scenario consists of seven parts, which are entitled:

Genesis.
Spirit and matter.
From darkness to dawn.
Man-made gods.
How time moulds civilization
Harmony.
To all mankind.

¹⁾ The Philips pavilion at the 1958 Brussels World Fair, I. Y. Xenakis, The architectural design by Le Corbusier and Xenakis,
II. C. G. J. Vreedenburgh, The hyperbolic paraboloidal shell and its mechanical properties,
III. A. L. Bouma and F. K. Ligtenberg, Model tests for proving the construction of the pavilion,
IV. H. C. Duyster, Construction of the pavilion in pre-stressed concrete,
Philips tech. Rev. 20, 1-36, 1958/59 (No. 1).

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SEQUENCE N° II

D'ARGILE et D'ESPRIT

Durée 60", De 61" à 120"

Ambiance n° 8 Durée 15"

Au niveau du sol : tout bleu, outremer massif.

Un soleil rouge intense uni et net.

Ambiance générale noire.

Aux niveaux supérieurs du pavillon deux parties jaunes.

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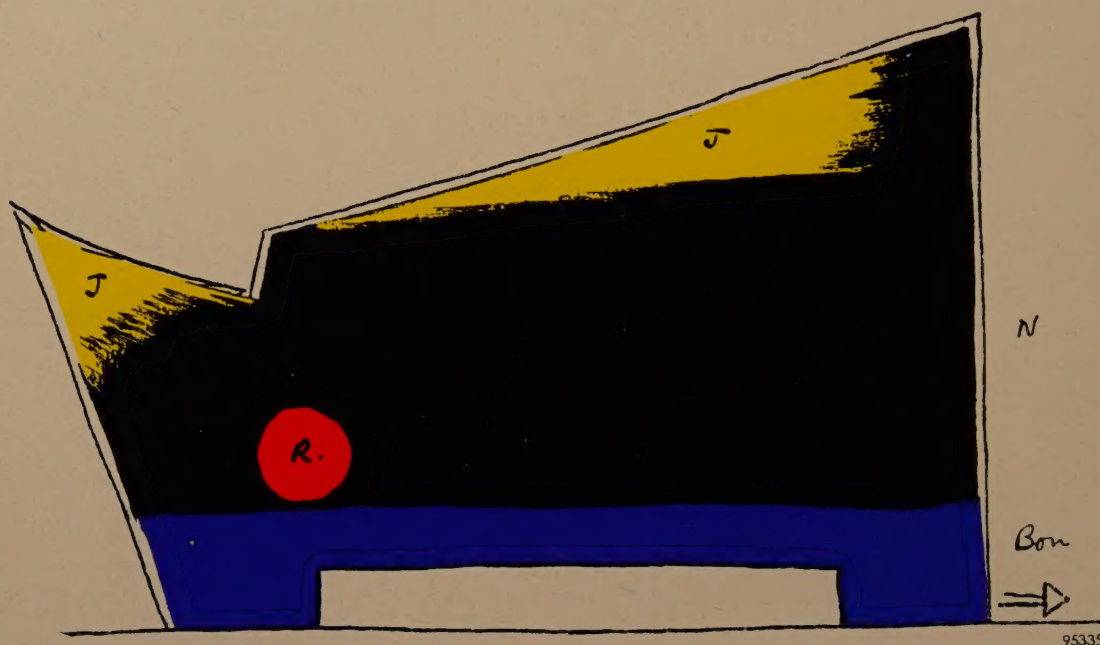


Fig. 1. Two pages of the scenario of Le Corbusier's "Electronic Poem". The page on the left indicates the colours of the "ambiances" in a given time interval; that on the right gives a time schedule, specifying the details of "écrans" and "tritrous".

Le Corbusier's scenario specifies what should be seen from second to second throughout the entire spectacle, which lasts 480 seconds. Two pages of the scenario are reproduced in *fig. 1*.

Le Corbusier distinguishes the following light effects:

- 1) "Ambiances", which are illuminated surfaces intended to conjure up a particular atmosphere. They are produced by projecting coloured light on to the pavilion walls; long bands of colour accentuate the peculiar shape of the building.
- 2) "Volumes", i.e. two figures suspended in space, the one a female figure, the other a space structure of metal tubes, and both treated with fluorescent paint. When irradiated with ultraviolet,

the one "volume" glows red, the other greenish blue.

- 3) "Ecrans": these are large areas of the wall on which are projected black-and-white pictures of human beings, animals and all kinds of objects (*fig. 2*). The presence at the same time of "ambiances" makes the black of the projections appear coloured.
- 4) "Tritrous". At certain parts of the wall around the "écrans", brilliantly coloured beams of light or figures in black and white are occasionally projected. This is done by means of a film strip each frame of which is entirely opaque except for three holes ("trois trous") in which the colours or figures are introduced.

[illegible]

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5) A red sun, a moon, stars and clouds can also be staged.

The technical arrangements made for producing these effects are relatively simple compared with the electro-acoustical equipment and the control system. Nevertheless the realization of the light effects presented certain difficulties. The greatest difficulty arose from the fact that the films and colours were to be projected from opposite sides of the enclosure simultaneously. This entailed a considerable amount of diffusely reflected light, which threatened to spoil the contrasts and the colour effects.

The problem was solved by 1) using the lowest practicable luminous intensities, so that not too much of the auditorium would be seen, 2) avoiding the use of large white surfaces, and 3) choosing suitable combinations of colours. Complementary colours, additively mixed, yield white or at least

colours that are less saturated, that is to say less striking. The answer to that was to use only non-complementary colours, such as green and blue, red and yellow, violet and red.

These measures demanded close cooperation between Le Corbusier and film-producer Agostini in regard to the prescribed pictures, and between the illumination engineers, whose task it was to decide on the kinds of light-sources and colour filters to be used. These questions were largely settled at the last moment by experiment in the pavilion itself.

The choice fell on the following light-sources:

a) Four large cinema-projectors, two of them for the pictures and two for the "tritrans". The "tritrans" beams are directed on to the required places by means of mirrors fixed at the appropriate angles. Synchronism between the projectors is assured by means of magslips.

b) Four projection lanterns, each with a 3 kW in-



Fig. 2. An "écran" (picture projected on the wall). The small rectangles are loudspeakers, of which there are 350 in the pavilion.

candescence lamp and a large rotary disc fitted with eight slides or colour filters for the "ambiances". Servo motors rotate the disc into the required position.

c) Two projection lanterns as under b) for producing cloud effects.

d) Two projection lanterns each with a 1 kW

incandescent lamp, for projecting a sun and a moon.

e) Six spotlights, each with a 500 W incandescent lamp, for projecting the colour "ambiances" onto the walls.

f) Four reflectors, each fitted with a 125 W high-pressure mercury-vapour lamp whose bulb trans-

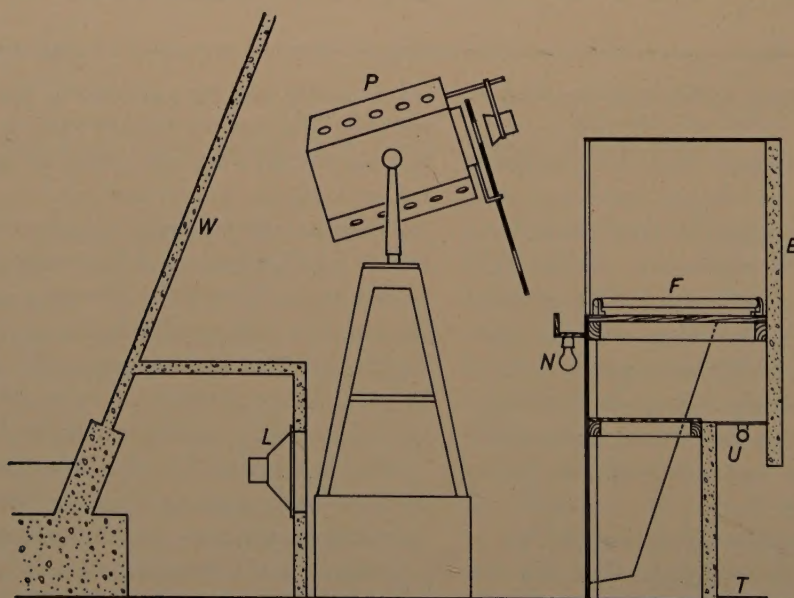


Fig. 3. Vertical section through the space behind the balustrade. *W* concrete wall of pavilion. *L* low-note loudspeaker in one wall of closed concrete cabinet. *P* projection lantern. *F* white and coloured fluorescent lamps. *N* lamp for emergency lighting. *U* tubular ultra-violet lamp for exciting floor-tiles *T* into fluorescence. *B* balustrade.

mits ultra-violet but hardly any visible light. Two of them excite red fluorescence on the female figure, the other two greenish-blue fluorescence on the space structure.

g) Fifty "stars", formed by incandescent 5 W

(40 W), in white, red, yellow, green and blue. These lamps are used to light the entire auditorium or, with reduced luminous flux, to create the illusion of a bright horizon. The lamps are accordingly mounted behind a six feet high

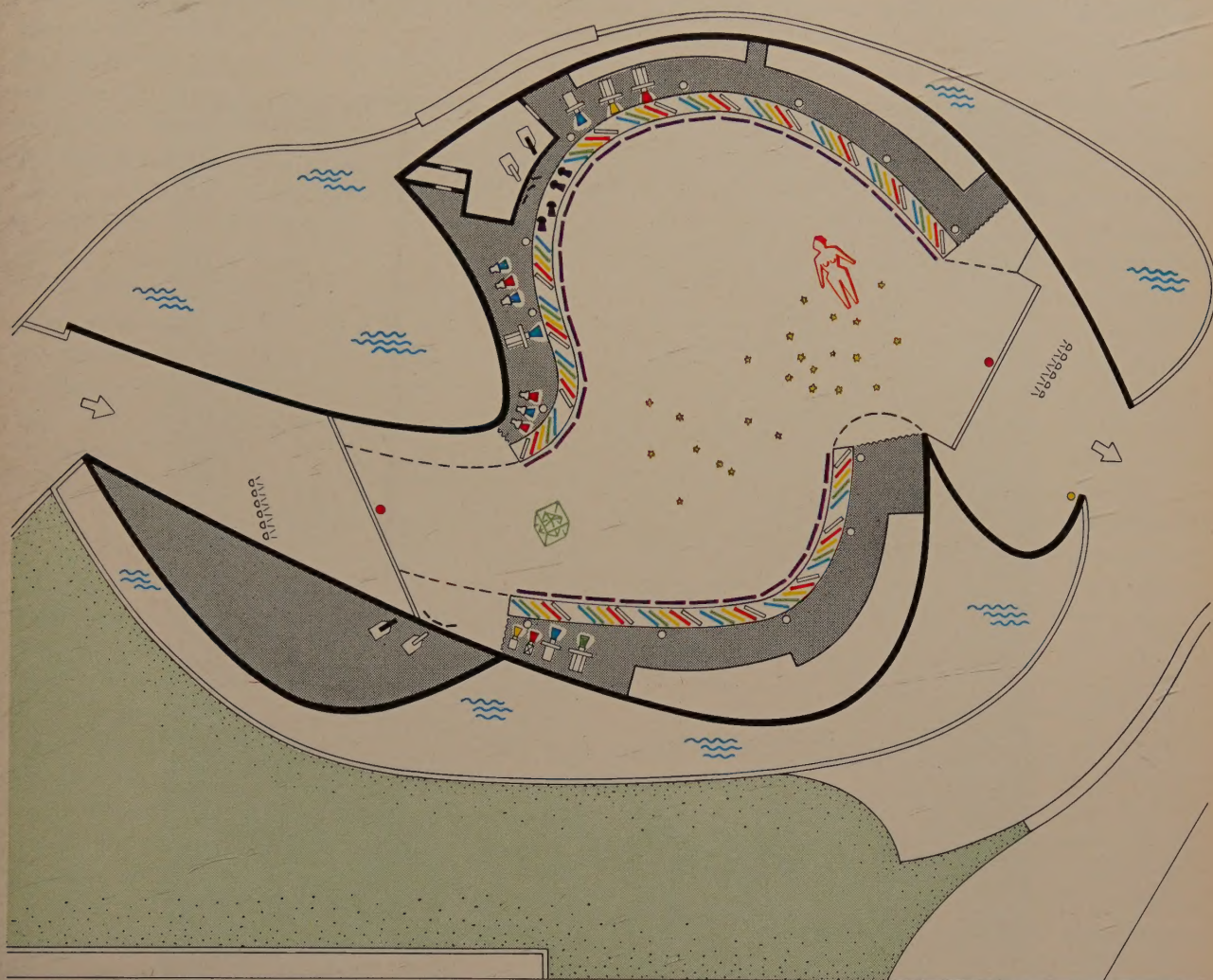


Fig. 4. Plan of the Philips pavilion with schematic indication of the light-sources.

bulbs scattered over the upper surface of the walls. A twinkling effect is produced by switching the bulbs on and off irregularly (by means of telephone selectors).

h) Forty groups of five fluorescent "TL" M lamps

balustrade which follows the contour of the floor (see vertical section in *fig. 3* and plan in *fig. 4*). The projectors are also mounted behind this balustrade (*fig. 5*).

The brightness of the fluorescent lamps behind the

balustrade is controlled by means of thyratrons²). The thyatron control-pulses are phase-shifted by the intermediary of potentiometers driven by servo motors. The latter are in turn driven by control signals recorded on a magnetic tape. Further particulars of the control system — which comprises a

lamps, burning continuously above the entrance and exit, and of several white bulbs that light up automatically in the event of a mains failure. To meet the contingency of panic amongst the audience, emergency lighting can be switched on from the control desk. Furthermore, bluish-white fluorescent



Fig. 5. Glimpse behind the balustrade. Some of the variously coloured fluorescent lamps can be seen and, behind them, an array of projection lanterns with filters for the "ambiances". Above, on the wall, a number of high-note loudspeakers. Bottom right a low-note loudspeaker.

great deal more — are given in the last part of this article.

The above projection and lighting installations serve exclusively for producing the light effects required for the performance of the spectacle. We shall now touch on the arrangements made for *utility lighting* in the pavilion (see fig. 4).

First, the regulation safety lighting is run on a 60 V battery and consists of two red incandescent

tiles are fitted in the floor around the whole length of the balustrade to prevent people knocking themselves upon entering the darkened pavilion. The tiles are excited into fluorescence by 45 tubular ultra-violet lamps of 40 W. In addition, six 40 W "TL" lamps concealed above the entrance on the outside produce a decorative effect at night, particularly since they are yellow fluorescing and hence provide a striking contrast to the blue mercury-vapour light which floods the exterior of the pavilion (16 high-pressure mercury-vapour lamps, each of 250 W).

²) K. W. Hess and F. H. de Jong, Controlling the luminous intensity of fluorescent lamps with the aid of relay valves, Philips tech. Rev. 12, 83-93, 1950/51.

B. THE SOUND EFFECTS

by W. TAK.

Acoustical science has long been familiar with various effects that influence our perception of sound. Reverberation and echo are the best known of these phenomena. Without the agency of visual perception, they provide us with an impression, learnt by experience, of our surroundings. Reverberation and echo can also be created artificially, and in such a way that the listeners' acoustical impression of their surroundings is made to differ appreciably from their visual impression³). This discrepancy between seeing and hearing can evoke in an audience the sensation of experiencing something startlingly unexpected. With such possibilities electro-acoustics may well contribute something to the development of music.

Equally remarkable effects can be obtained with the technique of stereophony⁴): sounds are heard to issue from a direction where there is no sound-source, and by means of purely electrical manipulations the impression of a moving sound-source is created without there being, in fact, any movement of the source.

These effects were in mind when, at the end of 1956, we discussed with Le Corbusier the sound effects for his "Electronic Poem". The listeners were to have the illusion that various sound-sources were in motion around them, rising and falling, coming together and moving apart again, and moreover the space in which this took place was to seem at one instant to be narrow and "dry", and at another to seem like a cathedral.

At that stage of the plans the music which Edgar Varèse was to compose was still quite literally music of the future. All that was known was that it would be mainly *electronic music*, in the connotation given to the term in recent years; that is to say, music whose actual composition is based on the use of electronic devices. As explained in an earlier article in this Review, electronic devices can be employed to produce entirely novel musical sounds⁵). Although we were as yet unaware of the composer's wishes in the matter, we were certain that stereo-

phony and artificial reverberation would turn out to be important elements of the "Electronic Poem". That was enough to enable us to plan in advance the general scheme of the requisite equipment.

It was our opinion that the simultaneous perception of three sound patterns issuing from or traveling in different directions would constitute a completely new experience. For this reason we decided there and then to develop and build apparatus that would be capable of reproducing the composition via three independent channels. We calculated that it would be possible to achieve the required electro-acoustical effects with the aid of a three-track magnetic tape. Each of the three playback heads would feed, via amplifiers, numerous loudspeakers. On another magnetic tape, having a large number of tracks, control signals would be recorded, some for determining which loudspeakers should be on at each moment, and others for controlling the light effects.

When these preparations were far enough advanced, Varèse came to Eindhoven to "produce" his composition with our cooperation. For this purpose a specially equipped studio was provided. In composing, Varèse concentrated primarily on the character of the tonal pattern, and for the most part left us to decide on the "intonation" (the distribution of the sound over the loudspeakers, i.e. the spatial effect). The composition is characterized by an extraordinary wealth of sounds, the realization of which often involved considerable difficulties⁶). The deficiency of language in this field, the lack of words to express what is intended, was keenly felt. Varèse frequently indicated his wishes by such expressions as "more nasal", "less biting", "more rasping", and it was our job to meet his wishes as well as possible by means of filters, mixers and frequency-shifting circuits. To define the necessary operations we had to resort repeatedly to onomatopœic words, such as "wow wow", "poowhip" "tick tock", "whoop" and "choochah". It should be mentioned, incidentally, that the recording was not limited to electronically produced signals but also included "concrete" sounds picked up by the microphone, for example piano chords, chimes of bells, choral and solo singing, drum beats and workshop noises, which, after electrical treatment (with filters, etc), were also recorded on tape.

³) Improving the acoustic properties of a hall by "stereo reverberation" (R. Vermeulen, Philips tech. Rev. 17, 258-266, 1955/56) is also based on bringing about a difference between the "acoustic" environment and the real one. In this case, however, the difference should be such that the listeners are unaware of it.

⁴) See, for example Philips tech. Rev. 17, 173 *et seq.*, 1955/56.

⁵) H. Badings and J. W. de Bruyn, Electronic music, Philips tech. Rev. 19, 191-201, 1957/58 (No. 6).

⁶) We are greatly indebted to Mr J. W. de Bruyn for solving many of these difficulties.

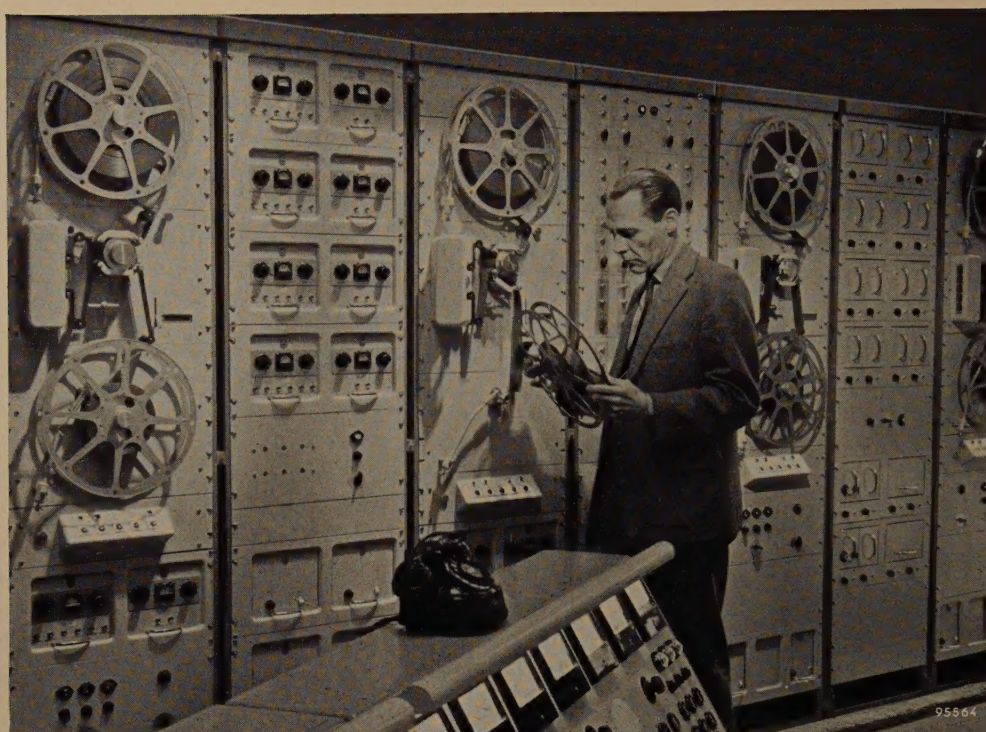


Fig. 6. Special playback machines for the 3-track tape carrying the electronic music by Varèse, and for the 15-track control tape. Two of the four machines are used for each performance.

The recording on the three-track tape was made so as to leave tracks *II* and *III* specifically for reverberation and stereophonic effects. For the purposes of stereophony, compact groups of loudspeakers would be necessary, e.g. one group above the entrance and another above the exit. However, it was not to be expected that everyone in the audience (the pavilion accommodates about 500) would receive a distinct stereophonic impression unless other, more dispersed, groups of loudspeakers were introduced, and the individual loudspeakers switched on and off successively in such a way that the sound-source would describe, as it were, set paths. This system of "sound routes" (*routes de son*) was therefore adopted and has proved successful.

After the composition had been completely built up on the three-track tape, a start could be made on working out a "control score". On a second magnetic tape — containing no fewer than 15 tracks — the control signals were recorded which were to assure the required distribution of the composition over the various loudspeakers. This was work calling for the most scrupulous accuracy, for every control signal had to appear or disappear at precisely the right moment. Further particulars of the control system are given in the next section.

Finally, a few words about the electro-acoustical equipment installed in the pavilion.

Both the sound tape and the control tape have

the same width (35 mm) and the same perforation as normal cinematographic film. The perforation is important with a view to the synchronous running of the two tapes. Each tape is scanned separately in a special magnetic playback machine (one with 3 and the other with 15 playback heads) and driven by a rapid-starting synchronous motor. With the exception of the loudspeakers, the entire installation is duplicated, which means that there are four playback machines in the control room (fig. 6).

There are altogether 350 loudspeakers. Twenty-five are low-note loudspeakers fitted in one of the walls of the otherwise completely enclosed concrete cabinets ⁷⁾ behind the balustrade (see figs. 3 and 5). The efficiency of these loudspeakers is particularly high (20%). The other loudspeakers (efficiency 5%) are mounted partly in compact groups (above the entrance and exit) and partly in dispersed groups (figs. 2 and 5), for example in "sound routes" along the shell ribs and horizontally. Over a complex relay system the loudspeakers are fed from 20 amplifiers, each with an output of 120 W. Six of the amplifiers derive their input signal from sound track *I*, eight from track *II* and six from track *III*. By means of special signals the inputs of the various amplifier groups can be interchanged, allowing a free choice of the path along which a certain sound is to be reproduced.

⁷⁾ See Philips tech. Rev. 18, 310 *et seq.*, 1956/57.

C. THE ELECTRONIC CONTROL SYSTEM

by S. L. de BRUIN.

As mentioned in the foregoing, the light and sound effects of the "Electronic Poem" are controlled by signals recorded on a magnetic tape. A simplified block diagram of the installation is shown in *fig. 7*. The control desks from which the duplicated installation is operated can be seen in *fig. 8*.

On *each* of the 15 tracks on the control tape, twelve control signals can be recorded, each with a fixed amplitude and a fixed frequency which differs from one signal channel to another. There are thus altogether $15 \times 12 = 180$ control channels available. The lowest frequency is 900 c/s (this fairly high value was chosen to exclude the risk of interference from hum), and the highest is 10 500 c/s. Neighbouring frequencies stand in a ratio of 1:1.25 to each other.

Each of the 15 playback heads delivers an output of 12 superimposed signals which is split into its components by selective amplifiers. These consist of two amplifying stages with tuned-circuit coupling and a relay. The relay closes only when the input signal in the first stage has the frequency to which the circuit is tuned. *Fig. 9* shows one of the consoles accommodating 48 of the total of 180 selective amplifiers.

Certain control channels are reserved for the light effects. Via some of these channels servo motors can be set in operation which rotate the required colour-filter or slide in front of the lens of the projection lanterns. Other channels control the thyristors, which regulate the luminous flux of certain

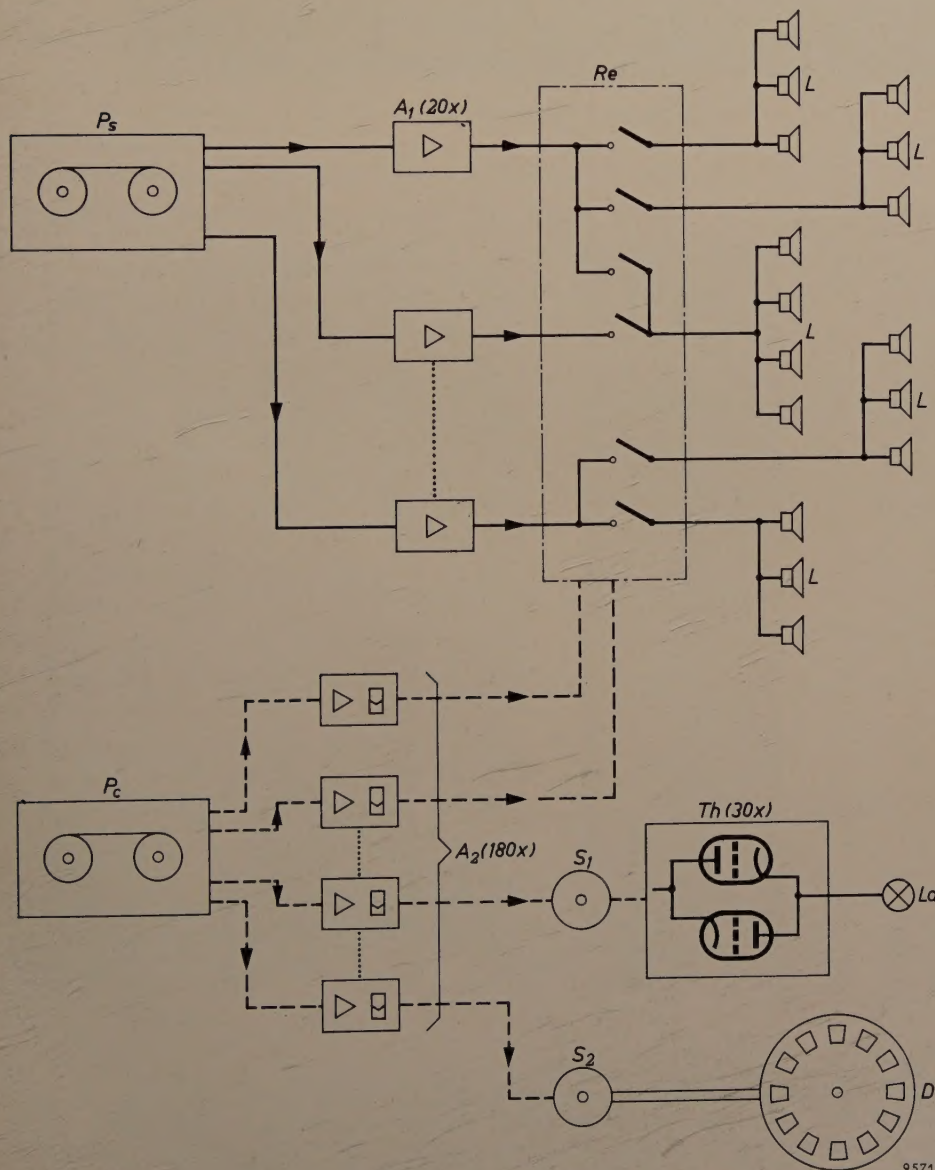


Fig. 7. Simplified diagram illustrating the principle of the installation for performing the "Electronic Poem". P_s playback machine with 3-track sound tape. A_1 amplifiers (20 in all) with 120 W power output. Re relays. L loudspeakers (350 in all). P_c playback machine with 15-track tape on which the control signals, with different frequencies, are recorded. A_2 selective control-signal amplifiers (180 in all); some of these control the loudspeaker relays Re , others the groups of servo motors S_1 and S_2 . The motors S_1 change the phase of the ignition pulses in the thyristor control-devices Th (30 in all), thereby varying the luminous flux of the lamps La . The servo motors S_2 rotate discs D which contain slides or colour filters.

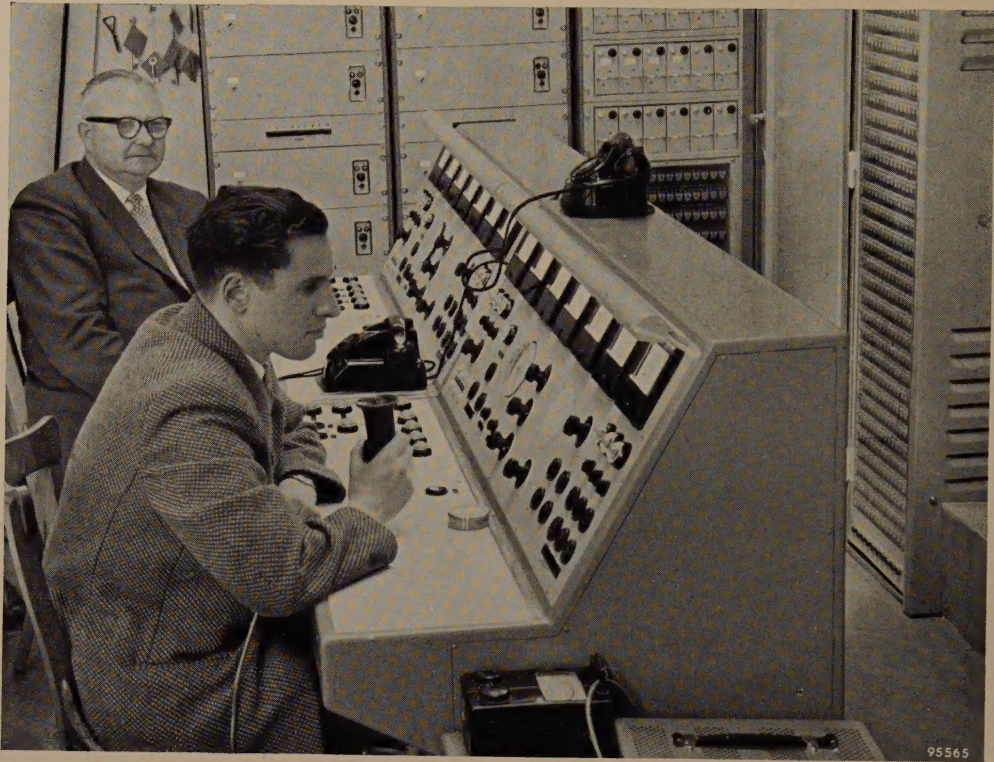


Fig. 8. The two identical control desks.

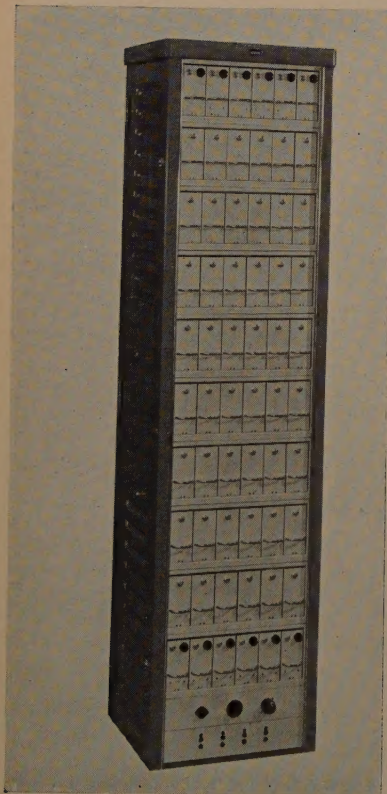


Fig. 9. Rack containing 48 of the 180 selective control-signal amplifiers.

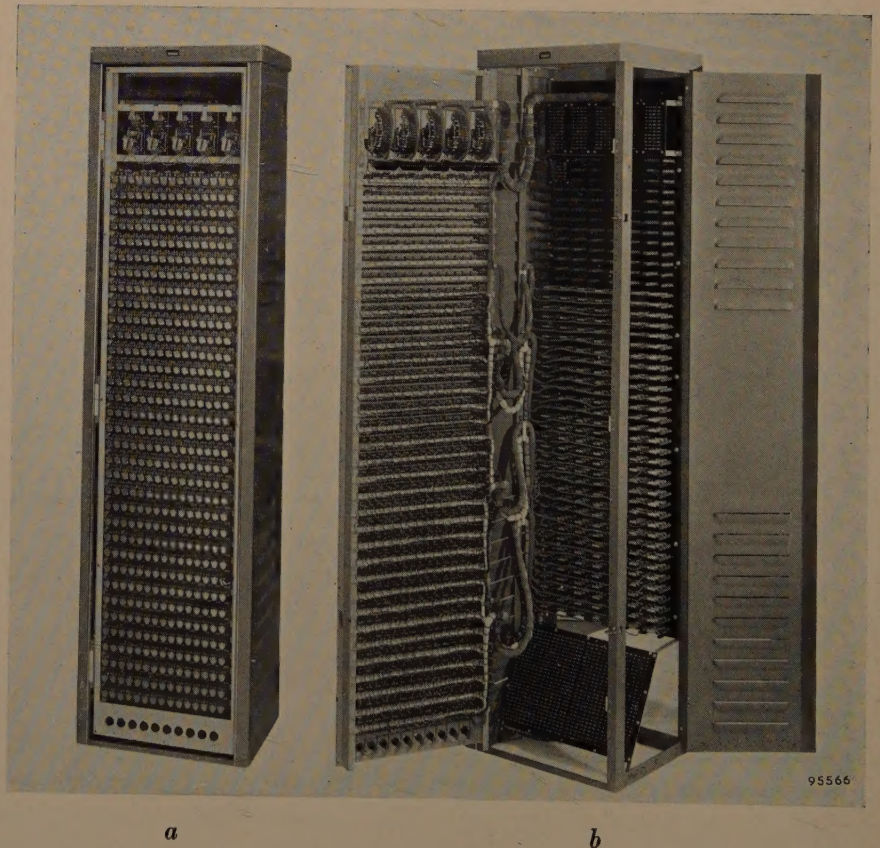


Fig. 10. Rack housing the loudspeaker relay system (*Re* in fig. 7), *a*) closed, *b*) opened. In *b*) can be seen the contact fields at which the loudspeakers and the amplifier outputs terminate. The five rotary selectors for the "sound routes" are visible at the top of the rack.

light-sources²⁾. This is also done by the intermediary of servo motors. In this case they turn potentiometers that vary the direct current through saturable reactors, thereby shifting the phase of the voltage pulses that ignite the thyratrons and hence changing the average thyatron current. Two thyratrons connected in anti-parallel are circuited in series with the light-source to be regulated. There are altogether 30 of these thyatron regulators.

The remaining control channels are used for the sound effects. They ensure that only those loudspeakers that are required are switched on at any given moment. The switching on and off of groups of loudspeakers is effected by relays (*Re* in fig. 7). A photograph of the relay rack is shown in fig. 10a. An interior view is seen in fig. 10b. The oblique panel below contains two fields of contacts; on the one field the amplifier outputs terminate, on the other the loudspeakers. Connections between any required loudspeakers and amplifiers (a maximum of 12 loudspeakers to one amplifier) are made or broken by the relays. These receive their energizing current via the contacts of the relays in the selective control-signal amplifiers.

The substantial number of relays needed for all this would have had to be very much larger if use had not been made of a circuit employing rectifier elements. The principle is illustrated in fig. 11 for three relays, but the circuit can be built for any number. Suppose that, with relays *A*, *B* and *C*, we wish at one instant to energize only *A*, at another instant both *A* and *B*, and sometimes *A*, *B* and *C* together. The first can be done by raising point 1 to a certain positive potential; *A* then closes and

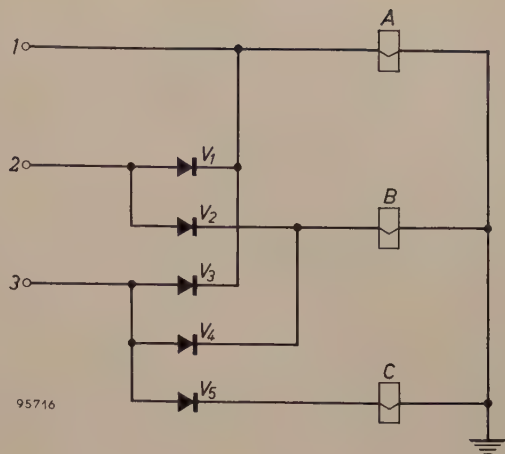


Fig. 11. Circuit with three relays, *A*, *B* and *C*, and rectifiers $V_1 \dots V_5$. A positive potential applied to points 1, 2 or 3 energizes respectively relay *A*, relays *A* and *B*, or relays *A*, *B* and *C*.

rectifiers V_1 and V_3 prevent current from reaching *B* and *C*. To energize both *A* and *B*, point 2 is given a positive potential, and rectifiers V_3 and V_4 prevent relay *C* from being energized. All three relays are actuated by giving a positive potential to point 3. Without the rectifiers, additional relays would be necessary, the contacts of which would make or break connections between the coils of *A*, *B* and *C*.

Reference was made above to the system of "sound routes" with which the illusion of a moving sound-source is created. This effect is produced by switching on first, say, only the loudspeakers $L_{121} \dots L_{125}$ (arbitrary numbering), a moment later $L_{122} \dots L_{126}$, then $L_{123} \dots L_{127}$, and so on, ending with, say, $L_{141} \dots L_{145}$ (fig. 12). The rapid switching from one set of loudspeakers to another is

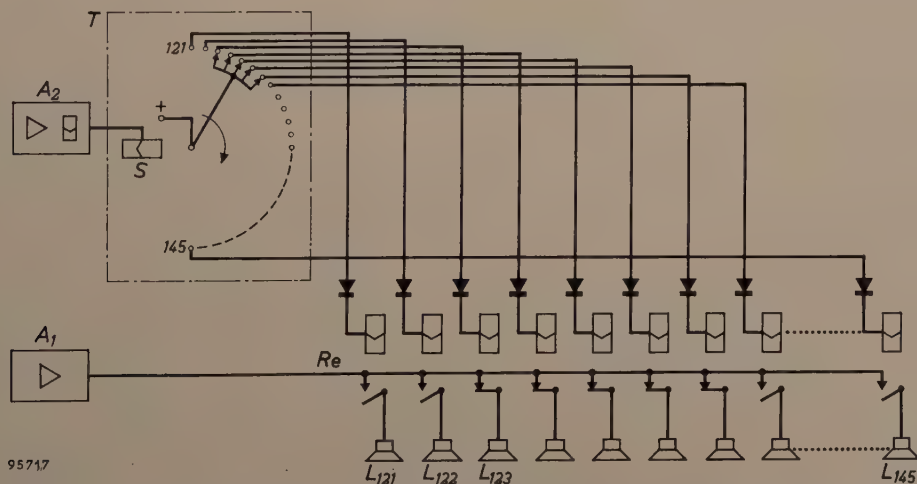


Fig. 12. Diagram of a "sound route", formed by loudspeakers $L_{121} \dots L_{145}$, which are fed from one of the power amplifiers A_1 via relay *Re*. The solenoid *S* of a uniselector *T* is energized by pulses from one of the control-signal amplifiers A_2 (see fig. 7). Five loudspeakers are always in operation at one time. At each selector step, one loudspeaker is switched in and another switched out.



Fig. 13

Fig. 13. Schematic representation of the recorded sounds and associated control signals in the time interval from 2 min 05 sec to 2 min 35 sec.

Below: the sounds recorded on the three tracks *I*, *II* and *III*. Above: the control signals present on tracks 7, 8, 9, 10 and 11 of the 15-track tape. The twelve frequency channels of each track are marked *a . . . l*.

In order, for example, to reproduce over loudspeaker group *A* the sound "tja-tju" recorded on track *I*, a control signal appears at the right moment in channel 8*g* and switches *A* on and off again. In the same way a control signal in channel 9*j* ensures that the two percussion sounds on track *II* are reproduced over loudspeaker group *U*.

The sound entitled "whistle finger" on track *I* is to be reproduced by the loudspeakers forming the "sound route" *O*. First, then, there must be a control signal in channel 9*b* for ensuring that group *O* will be available during the required time. As a rule this group reproduces sounds from track *II*. "Whistle finger", however, is on track *I*, so that a control signal is needed which will switch over the input of the amplifier for

group *O* from track *II* to track *I* at the appropriate moment; this signal appears in channel 11*j*. When the above two signals are present, a train of 60 pulses (at a frequency of 4 per second) begins in channel 9*k* and these pulses cause one of the rotary selectors (*III*) to make a full revolution. This results in the functioning of "sound route" *O*. The selector must now be returned to its starting position, and this is done by the signal "reset *k III*" in channel 9*a*. To ensure that the selector returns exactly to its starting position, even if it has covered too few steps, the number of pulses in channel 9*k* is made somewhat larger than the number of selector positions (60 pulses, 52 positions). The simultaneous presence of these pulses and the reset signal prevents the selector from rotating beyond its starting position.

Reset signals also appear in channels 7*a*, 8*a*, 10*a* and 11*a* for returning selectors *V*, *IV*, *II* and *I* to their starting positions.

The control signals marked *L.t. II* and *L.t. III* cause the low-note loudspeakers (which form one group) to reproduce sound from tracks *II* and *III*, respectively.

effected by selectors as used in automatic telephone exchanges (*T* in fig. 12). These are operated by signals on the control tape which consist of short pulses with a maximum repetition frequency of 10 per second. The selectors are visible at the top of the rack shown in fig. 10*a* and *b*.

If the loudspeakers of the group concerned are disposed around the auditorium, "circulating sound" is produced by letting the selectors rotate completely several times. Special control signals return the selectors to their starting position after the completion of a "sound route".

Fig. 13 shows a diagrammatic representation of the sound with the associated control signals. The sounds on the three tracks during the interval represented are indicated below, while above can be seen the control signals required for these sounds to be reproduced via the appropriate loudspeakers. Some of the signals are explained in the caption; further details would exceed the scope of this article.

The "Electronic Poem" lasts eight minutes. Each performance is preceded by an introduction in

English, French and Dutch, which takes two minutes. Some twenty performances are given daily, alternately via the two installations, which amounts to many thousands of performances throughout the exhibition.

Summary. In the Philips pavilion at the Brussels World Fair performances are given of Le Corbusier's "Electronic Poem", which is a combination of light effects and electronic music involving extensive technical provisions.

The equipment for the light effects includes four large cinema projectors, eight projection lanterns, six spotlights, six ultra-violet lamps, fifty electric bulbs representing stars, and several hundreds of tubular fluorescent lamps in various colours. The luminous flux of most of these light-sources is variable by means of 30 thyatron control circuits.

The electro-acoustical installation comprises a special playback machine with a three-track perforated magnetic tape on which is recorded a composition by Edgar Varèse, 20 amplifiers each with an output of 120 W, 350 loudspeakers and an elaborate switching system with relays and telephone selectors.

The electronic control system employs a second playback machine, which scans a 15-track perforated magnetic tape. Twelve control signals differing in frequency are recorded on each track, so that 180 channels are available for controlling the light-and-sound effects in accordance with the scenario. Both playback machines are duplicated, with a view to providing a reserve as well as ensuring continuous performances.

NOISE

A GENERAL SURVEY

by K. S. KNOL *).

621.396.82

*In this inaugural address as Professor of electronics at the Eindhoven Technische Hogeschool on 17th December 1957, Dr. K. S. Knol reviewed the advances made in understanding the phenomena which give rise to reception disturbances in telecommunication engineering, and which are collectively called "noise". Among the aspects touched on were noise control and some applications of our present-day knowledge in pure scientific research and in industry. With Professor Knol's kind consent and cooperation we print below the principal contents of his address **), supplemented by a bibliography and some illustrations.*

When we try to tune our radio set to a far-distant or weak station we sometimes find that the programme does not come clearly and distinctly through the loudspeaker, but is drowned by crackling and hissing sounds. Particularly characteristic of such sounds are those heard from a frequency-modulated receiver when it is not exactly tuned in. Usually they disappear when the set is tuned to a powerful enough station.

Owners of a television set who attempt to watch a programme from a transmitter which is not within the range necessary to ensure good reception will be familiar with a sight on their screen more resembling a snowstorm than the scene transmitted from the studio.

Both phenomena — hissing and crackling from the loudspeaker and "snow" on the television screen — have the same causes, and in technical parlance the phenomena as well as the causes are collectively termed "noise".

Unattractive as the phenomena may be, the study of noise and with it the search for means of approaching as closely as possible to the natural limit to the amplification of signals, present the research worker with many fascinating problems.

The first problem that arises in this connection is: does the noise originate from inside or outside the receiver? The answer is that both may be the case. We shall first, very briefly, consider noise originating outside the receiver, that is to say noise caused by interfering signals which enter the set either via the aerial or through the cables of the electric mains. Next we shall deal in greater detail with the phenomena causing noise inside the set and which,

generally speaking, set a limit to the useful sensitivity of the receiver.

External sources of noise

Interfering signals from outside the set can come from near and afar. Those from nearby may originate in electrical equipment, such as motors, gas-discharge lamps, high-frequency heating apparatus and the ignition systems of cars and other vehicles in which electrical discharges occur. Such interference is known as "man-made noise", and can be suppressed at source.

Another kind of disturbance is to be sought somewhat farther from home, but is still very terrestrial in origin. A thunderstorm within a few miles of the receiver produces, with each lightning flash, a momentary crackling in the loudspeaker. This, however, is not classed as noise. The innumerable discharges from the electrical storms that rage almost without abatement around the earth, and particularly in the tropics, cause continuous crackling interference having the character of rustling. This interference is noticeable particularly on waves that are generally longer than about 10 metres, and which reach the set after travelling a very great distance via multiple reflections between the ionosphere and the earth's surface.

The third kind of noise that can penetrate our receivers from outside is extra-terrestrial in origin, coming to us from outer space: it is called "cosmic noise". Its study has led in latter years to the growth of an entirely new branch of science, radio-astronomy¹⁾, the rapid progress in which has made it a most valuable complement to classical astronomy. Recent results have supplemented in many respects the astronomer's picture of the cosmos. It should be noted, though, that the radiation predicted by Van de Hulst²⁾ — the 21 cm line of hydrogen — which has thrown much light on the structure of the

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**) The full text of the address (in Dutch) has been published by J. B. Wolters, Groningen-Djakarta.

galactic system in particular, cannot readily be classed as noise. The narrow bandwidth of this radiation makes it possible to demonstrate its presence in cosmic noise. It is so weak, however, that it cannot be distinguished from the noise of the receiver other than by special means, and its investigation is therefore primarily a noise problem. It would exceed the scope of this survey to go any deeper into this interesting object.

Internal sources of noise

Thermal noise

The study of the electrical fluctuation phenomena which, quite independent of outside phenomena, give rise to noise in the receiver, is relatively young, dating only from the beginning of this century. Before that time the electrical methods of observation were not sufficiently advanced to demonstrate these phenomena.

A continuous irregular movement of very small particles in a liquid had, however, already been noticed in 1827 by the English botanist Brown. A satisfactory explanation of this Brownian movement, as it is called, was not forthcoming until much later. It was found to be due to the thermal agitation of the liquid molecules, which collide with the larger particles, visible under a microscope, and so cause their random movement. In 1906 Einstein and, working independently, Smoluchowski, succeeded in calculating the mean-square velocity of a particle in random thermal movement³⁾, and found it to be proportional to the absolute temperature. Another significant step forward was taken when Smoluchowski predicted in 1912 that Brownian movement could cause spontaneous deflections of a galvanometer coil⁴⁾. It was not until 1925 that such deflections could be actually demonstrated. In that year Moll and Burger reported a method of making an extremely sensitive galvanometer with the aid of a "thermo-relay"⁵⁾. Their galvanometer was in fact so sensitive that it showed spontaneous deflections. They attributed these to microseismic disturbances. Ising⁶⁾ was able to show, however, that these fluctuations could be largely explained by Smoluchowski's theoretical predictions.

It is evident that spontaneous and completely random movements of a galvanometer coil, small though they may be, must set a limit to the useful sensitivity which a galvanometer can be given.

In 1912 there also appeared the still quoted thesis by De Haas-Lorentz⁷⁾ in which she studied, among other things, the effect of the thermal agitation of electrons in electrical networks, thereby continuing

a line of investigation already started by Einstein.

In 1928 Nyquist and Johnson demonstrated that the thermal agitation of the electrons in resistors used in amplifiers of electrical signals also set a limit to the useful sensitivity of the amplifiers⁸⁾. Using the laws of thermodynamics, Nyquist calculated the mean-square noise voltage, in a small frequency interval, occurring across a resistor. He found that this, too, was proportional to the absolute temperature T , and also to the value R of the resistance and the width Δf of the frequency interval, but *independent* of the frequency:

$$\overline{v^2} = 4kTR\Delta f,$$

k being Boltzmann's constant.

This result was subsequently confirmed from several quarters. In 1939, for example, Bakker and Heller⁹⁾ calculated the noise current in a wire short-circuiting a resistance, by summing the effect of the movements of all electrons in the resistance, thereby obtaining a result analogous to that of Nyquist.

In all these calculations it is assumed that the resistance is carrying no current, the only consideration being the thermal agitation of the electrons, i.e. thermal noise.

Shot effect

Another kind of fluctuation phenomena occurs when, although there is a continuous current of particles, the particles do not behave like a column of well-disciplined troops, marching in step, but move in more random fashion. We might compare the situation with a downpour of hailstones; if we were to count the number of hailstones falling on a given area in a given time, and we repeated the count at different intervals, we should find that the totals were not identical, but fluctuated around a certain average. The same applies, for example, to the electrons travelling from cathode to anode in a thermionic valve, or to the number of quanta impinging per unit area in an X-ray image intensifier. These fluctuations also give rise to noise. Schottky, who first discovered this kind of noise in 1918¹⁰⁾, called it "Schroteffekt" and in the English-speaking countries it has become known as "shot effect".

Schottky worked out a formula for the noise current in a saturated diode, showing that the mean square of the noise current, $\overline{i^2}$, is proportional to the direct current I_s through the diode, to the electronic charge e and to the frequency interval Δf , but again independent of the frequency itself. This brings out very clearly the corpuscular character of the electron current. Schottky's formula made it possible to determine from noise measurements the charge of a

single electron; this was the first instance of the use of noise measurements as an aid to physical research. The first attempt to measure the elementary charge in this way was made by Hartmann¹¹⁾, but the result was not satisfactory. For that reason Johnson¹²⁾ checked Schottky's calculations and found that somewhere a factor of 2 had been forgotten. The corrected formula was

$$i^2 = 2eI_s \Delta f,$$

but this was not entirely consonant with Hartmann's results. Later the measurements were performed with greater accuracy and results obtained for e that agreed with the values found by other means.

The saturated diode, for which Schottky devised his formula, is nowadays still in daily use as a noise standard in many laboratories (*fig. 1*).

It is a fortunate fact that in most cases the noise in thermionic valves turns out to be a lot less than that in a saturated diode. In the case of valves operating in the space-charge region, where a potential minimum occurs in front of the cathode, the noise current in the anode circuit is found to be greatly diminished. Calculations of the noise current under such conditions have been made by Schottky, Spence, Rack and North¹³⁾, and others.

Other causes of noise in thermionic valves

In more complicated valves — for example those with more than one grid, such as tetrodes, pentodes and hexodes — a further cause of noise can be the fluctuations due to the division of the electron current between two positive electrodes (partition noise)¹⁴⁾. The chance factor involved in the number of electrons striking or missing an electrode gives rise to fluctuations in the current to these electrodes.

In valves with secondary-emission electrodes an additional noise current is caused by random fluctuations in the number of secondary electrons liberated per primary electron¹⁵⁾.

Noise in semiconductors

What we have so far discussed really applies only to thermionic valves that are not operated at extreme frequencies. In the electronic devices based on semiconductors, such as the crystal detectors of early radio sets and the transistors of to-day, we might also, very broadly, distinguish between thermal noise and shot effect for a certain "intermediate" band of frequencies, i.e. frequencies that are not too high and not too low. This frequency band, however, is not the same as for thermionic valves, which is hardly surprising since in semiconductors the flow

of current is primarily a diffusion process at room temperature, the charge-carriers having a much lower velocity than in thermionic valves. Consequently the transit-times of the charge-carriers in semiconductors are a great deal longer and transit-time effects therefore play a part at much lower frequencies. In the case of recently-developed transistors, however, whereby use is made of an internal accelerating field, the influence of transit-time effects can be shifted to much higher frequencies.



Fig. 1. A modern standard noise-diode, as described in a forthcoming article by H. Groenijk in Philips tech. Rev. 20, 108-110, 1958/59 (No. 4).

Before dealing with the subject of transit-time effects, let us turn briefly to another and highly important type of noise, which plays a significant part in both thermionic valves and semiconductors, and which appears at very low frequencies.

Noise at very low frequencies

At very low frequencies the noise of thermionic valves and semiconductors is found to become strikingly high. In this case the mean-square noise current in a narrow frequency interval is usually inversely proportional to the frequency:

$$i^2 \propto \frac{1}{f}.$$

The first types of point-contact transistors showed this kind of noise to a particularly marked degree.

In 1926 Schottky published a theory of this noise effect in thermionic valves¹⁶⁾, which was not, however, capable of explaining all the phenomena. Schottky assumed the occurrence on the cathode of

emission centres which, for a certain time, their lifetime, emit a strong current. On the basis of a certain specific lifetime, he calculated the noise spectrum and found that at very low frequencies \bar{i}^2 should be constant, while at high frequencies it should decrease in approximately inverse proportion to the square of the frequency:

$$\bar{i}^2 = \text{const. (f low),}$$

$$\bar{i}^2 \propto \frac{1}{f^2} \quad (f \text{ high}).$$

The transition between these two regions would be expected to be at a frequency for which the period of the oscillation is of the same order of magnitude as the life of an emission centre. In practice it has almost invariably been found that \bar{i}^2 is still about inversely proportional to f down to extremely low frequencies. The same applies to semiconductors and resistors made of very thin films of metal or carbon through which a current is passed. In fact Rollin and Templeton have shown¹⁷⁾ that with a resistor of such a kind this noise spectrum extends to frequencies of the order of 10^{-4} c/s, i.e. about 1 cycle per hour (fig. 2).

From Schottky's theory also followed a law for the suppression of this noise by the space charge. Measurements proved, however, that the noise suppression was quite different from that predicted by Schottky, and therefore the theory of emission centres had to be abandoned.

It seems likely that variable processes must play some part in cases where the noise is dependent on frequency. A mechanism depending on a single-valued lifetime evidently cannot give an adequate explanation of the spectrum found. It might be possible, however, to explain it by a superposition of processes of widely divergent lifetimes. Recently the existence of such many-valued lifetimes has been demonstrated in the case of energy states on the surface of germanium, by means having nothing to do with noise measurements. For this reason there is a tendency at present to assume that the spectrum in question is caused by a modulation process due to energy states of this nature. Spectra are also found, incidentally, which differ in varying degrees from the type in which \bar{i}^2 is inversely proportional to the frequency.

At all events, the nature of this low-frequency noise is extremely complex and manifests itself in dissimilar ways with different materials. Van der Ziel and co-workers have latterly done important work on low-frequency noise in thermionic valves¹⁸⁾. One particularly interesting result is that their noise

measurements have provided indications supporting the pore-conduction mechanism, postulated by Loosjes and Vink¹⁹⁾, in the oxide cathode. This is a striking example of how noise investigations can be used to gain an insight into other physical phenomena.

Noise at very high frequencies

The causes of noise that dominate at low frequencies have scarcely any influence at very high

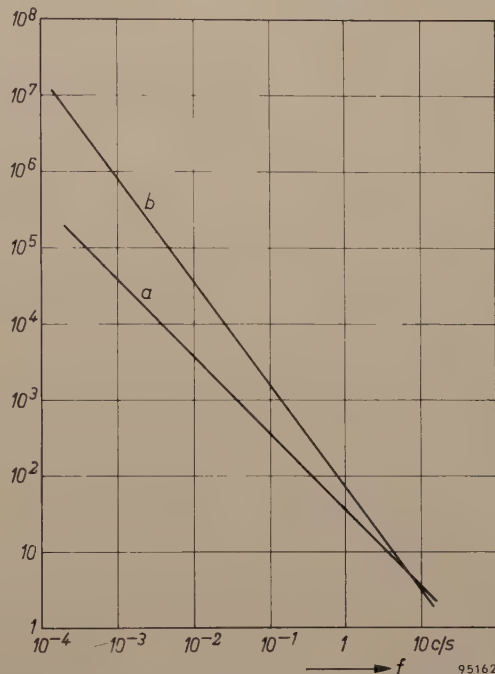


Fig. 2. Relative value of the mean-square noise current per unit bandwidth, as measured at very low frequencies by Rollin and Templeton¹⁷⁾. For curve *a*, measured on carbon resistors, $\bar{i}^2/\Delta f$ is inversely proportional to the frequency f ; for curve *b*, measured on a germanium wire of 10 kΩ resistance, it is inversely proportional to $f^{1.35}$.

frequencies. At extremely high frequencies one would even expect a decrease in the thermal noise, as a result of the effects of the quantized nature of the electromagnetic field. At normal ambient temperatures the wavelengths at which quantum effects become noticeable are so short — a few hundredths of a millimetre — that there seems no likelihood at present of their assuming any practical importance. Nevertheless, it should be possible to demonstrate these effects in the laboratory, where the means are available for producing extremely low temperatures and the necessary high frequencies in the microwave region. One would like to see cooperation to this end between specialists in microwave and low-temperature techniques.

Noise phenomena are, however, noticeably influenced at high frequencies by the electron transit-

time effects to which we have already referred. This is manifested in two ways:

- 1) changes take place in the shot-effect noise current, already present at low frequencies;
- 2) phenomena appear that are not perceived at low frequencies.

To ascertain the effect of electron transit-times on the noise also present at low frequencies, let us consider a diode having a potential minimum due to space charge. No theory yet exists that accounts in all details for the fluctuations in the anode current for such a case. That being so, one has to resort to an idealized model, assuming that in or just beyond the potential minimum an electron beam emerges with a certain average speed, and that the density and velocity of the electrons show fluctuations around their mean values. But if we try to calculate these fluctuations and their mutual correlation in the potential minimum, we come up against a theoretical problem of considerable difficulty. One might indeed expect, at first sight, that the fluctuations in the number of electrons passing the potential minimum would be related to the fluctuations in the mean velocity of the electrons. However, when we go deeper into the matter we find that with the existing theories it is impossible to demonstrate any correlation. For this reason most investigators take the view that none exists. A mathematical approach has recently been published by Tien *et al.*²⁰⁾ in America, who have studied a much more intricate model. With the aid of a modern computer they have attempted to calculate the paths and the behaviour of numerous relatively small mutually-interacting groups of random numbers of electrons emitted successively by the cathode at identical, very short intervals. As can easily be imagined, this was a task of intimidating proportions. The result of the calculations was extraordinarily interesting as regards the expected noise behaviour of travelling-wave tubes, but it yielded no correlation between density and velocity fluctuations.

On the basis of the density and velocity fluctuations in the potential minimum as derived from the theory applicable to low frequencies, one can calculate with the aid of the existing transit-time theories the fluctuations at a position farther on in the beam or in the external circuit. Mention should be made here of the important work by Cutler and Quate²¹⁾, who have shown that the noise in such electron tubes exhibits maxima and minima along the beam (fig. 3). Their work led to the development of low-noise travelling-wave tubes.

In simple diodes and triodes the transit-times are

usually not long enough to allow the formation of waves. The gradually evolved theory, as finally formulated by Llewellyn and Peterson²²⁾, is sufficient in their case.

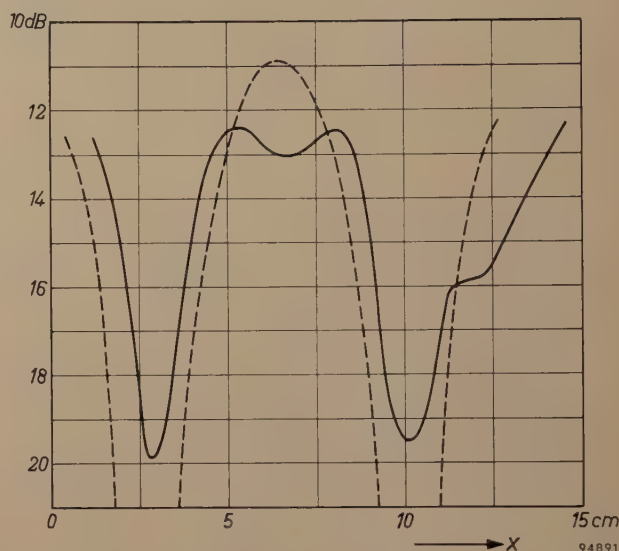


Fig. 3. Mean-square noise current in the electron beam of a travelling-wave tube, as a function of distance x from the anode (after Cutler and Quate²¹⁾). On the ordinate is plotted the number of decibels below the level of the shot effect. The solid curve represents measured values, the broken curve calculated values. Maxima and minima appear along the length of the beam.

Among the phenomena that only become perceptible at high frequencies are *total-emission noise* and *induced grid noise*²³⁾. The first is due to the many electrons that return to the cathode without having reached the potential minimum. Practically all emitted electrons are involved in this process, hence the name total-emission noise. They make, as it were, brief excursions into the space between the cathode and the next electrode, and then pass directly back to the cathode. Each electron induces in the cathode circuit two equal but opposite tiny current pulses, which are only slightly displaced in time. The noise current so induced in the cathode circuit is therefore noticeable only at extremely high frequencies.

Induced grid noise is due to the fact that every electron travelling from cathode to anode via a grid induces in the grid circuit two almost identical and opposite current pulses, again slightly displaced in time. At high frequencies the pulses of all electrons together give rise to a perceptible noise current in the grid circuit. Since this current owes its existence to the electron current through the valve, there should be complete correlation with the noise current in the anode circuit; it should therefore provide us with a means of reducing the anode noise-current.

Strutt and Van der Ziel have indicated how such a noise reduction might be accomplished²⁴). Unfortunately experiments show that the correlation between induced-grid noise and anode noise-current is not as was expected. Various laboratories are working on the problems still to be solved in this connection, the frequencies involved ranging from several hundreds to several thousands of megacycles.

The noise characteristics of radio receivers

Now that we are familiar with some of the causes of noise in the elements of a receiver of electromagnetic waves, let us see how we should define the sensitivity of a receiver in the light of these considerations. For clear undisturbed reception of a radio or television programme, the ratio of the signal power to the noise power of the sound or picture is obviously important; it is referred to simply as signal-to-noise ratio.

Together with the signal the aerial also receives noise, the amount of which may vary according to the aerial system. To provide a basis of comparison it is assumed that the noise received corresponds to the thermal noise produced at room temperature in a normal resistance equal to the radiation resistance of the aerial.

To characterize the noise behaviour of a receiver, Friis²⁵) introduced the term "noise factor", which denotes the noise-to-signal ratio — i.e. the reciprocal of the signal-to-noise ratio — at the output of the amplifier divided by that of the aerial. This definition, seemingly simple and unambiguous, must nevertheless be used with caution in practice. The reason is that the most favourable noise factor is usually obtained not when the amplification is maximum, but when the aerial is matched to the first amplifier valve, without the transmission of energy being a maximum.

The radio designer intent on building a receiver of optimum sensitivity must of course strive to keep the noise factor as small as possible. To this end he needs data from the valve manufacturer that will enable him to design a circuit which, with due regard to other requirements, such as the desired bandwidth, etc., will have the lowest possible noise factor. At the same time he wants valves that produce little noise.

The valve manufacturer, then, knows what is required of him. The demand for valve data can be readily complied with. There are various methods of specifying the noise characteristics of an amplifier valve, transistor or crystal diode without entering directly into their mode of operation. We shall

mention here a very simple method first proposed by Becking²⁶). He showed that the contribution to the noise of a fourpole can be fully described by imagining that a noise-voltage source and a noise-current source, which may be correlated, are connected at the input of the fourpole, which itself is thought of as being noise-free. The noise behaviour of the fourpole can then be defined in terms of four easily measured quantities, knowledge of which makes it possible to calculate the noise factor of each input circuit and, conversely, to find that circuit which gives the lowest noise factor. This is sufficient for the radio designer's needs.

The man engaged on valve research, however, is not content with the measurements of noise characteristics alone, but wishes to correlate their values with the physical phenomena inside the valve. This presents various problems that add greatly to the interest of noise investigations. We have already touched on the problem of the fluctuations in the velocity and density of the electrons in the potential minimum, and on the imperfect correlation between induced grid noise and anode noise. For total-emission noise, too, there exists as yet no theory capable of explaining the experiments quantitatively, while as regards semiconductors there are still many unknown factors to be resolved, which are essentially analogous to those of valves.

In every individual case we can try to ascertain the minimum noise factor attainable. The results will vary widely in different frequency bands. The value 1 can never be reached, for this would mean that the amplifier made no contribution to the noise. This, of course, represents the unattainable ideal. In many cases there is no need to make great efforts to reduce the amplifier noise since the noise received by the aerial on normal broadcast wavebands is so high that there is little point in making the receiver very sensitive. For short-wave and television reception, on the other hand, the noise factor can still be usefully reduced, while as regards radar and microwave links for telephony and television, noise remains a stumbling block.

On centimetric waves the lowest noise factors have so far been achieved with travelling-wave tubes. A very low noise factor is difficult to obtain with a triode because of the high level of grid noise and because of the limited noise suppression at present attainable owing to certain still not entirely explained sources of noise in triodes. Efforts are now being made, by further refining an already extremely refined technique and by adopting measures derived from deeper theoretical considerations, to fabricate triodes having a very low noise factor.

The lowest possible noise factor is of paramount importance in equipment used for measuring cosmic noise¹). For this purpose there is a constant demand for valves and other components of lower inherent noise. One might think in this connection of using a very low-temperature gas or solid, in view of the great reduction of the contribution due to thermal noise at low temperatures. Various possibilities are being studied at the present time. It is quite conceivable, for example, that semiconductors might be made having low-temperature characteristics analogous to those of silicon and germanium at room temperatures.

An extraordinarily interesting development has been taking place in recent years, principally in America, whence the news of "microwave amplification by stimulated emission of radiation", abbreviated to "maser", was disclosed to a surprised world some years ago. The first publication on the maser (*fig. 4*) came in 1955 from Townes and co-workers²⁷), who had been working on a gas. About a year later Bloembergen demonstrated that the same principle was applicable to solids²⁸). Reports from Russia, too, indicate that similar principles are being applied there with success.

From calculations which seem to be reliable and from preliminary measurements, it appears that very low noise factors are to be achieved by the maser principle. Its application to solids calls for extremely low temperatures. Masers involve rather extensive set-ups, so that it does not look as if they will be put to general use for the time being. For special purposes, however, such as the measurement of cosmic noise, it is possible that the maser will find practical application in the not too distant future²⁹).

It is gratifying that a new development of such significance, springing from pure scientific research, should immediately arouse universal interest and give rise to active investigations in laboratories all over the world.

Noise research in the study of physical processes

We have hitherto been discussing noise chiefly as something that limits the sensitivity of receivers, that is to say, regarding it from the standpoint of radio and valve designers. In passing we have referred to the results of noise research as an aid to the understanding of certain physical processes, but in most cases we have been concerned with noise studied as an end in itself. Quite often, however, noise research can be used as a *means* of studying physical processes. We have already mentioned

radio-astronomy, the determination of the elementary charge and pore-conduction in the oxide cathode. We can also include under this head the work of Van Vliet³⁰), who has been able to draw some important conclusions on electron transitions in single crystals of cadmium sulphide.

Another example is Dahlke's application of noise research for investigating the emission properties of cathodes³¹). This line of investigation is of considerable importance to the factory laboratory, showing as it has done that the effect of inadequate emission

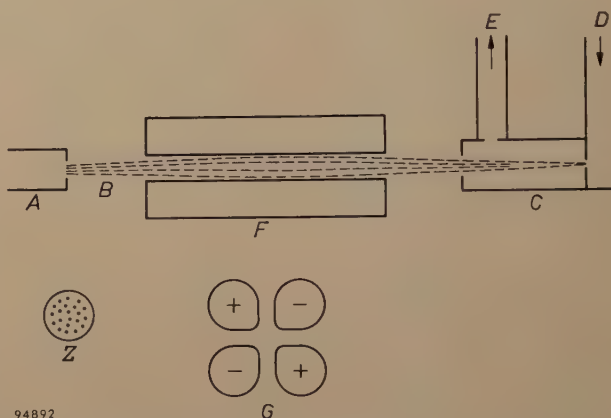


Fig. 4. Simplified diagram of a "maser" arrangement ("microwave amplification by stimulated emission of radiation"; see, for example, W. H. Culver, *The maser*, Science **126**, 810-814, 1957). A beam *B* of NH_3 molecules emerges from a source *A* via numerous fine channels (end view *Z*). *F* is an electrostatic electrode system (transverse section *G*) which selectively focuses the molecules present, causing those in the upper energy-level of a certain transition to converge at *C*. *C* is a cavity resonator in which these molecules, in their transition to the lower level, give up energy to microwaves of suitable frequency fed in through waveguide *D* ("stimulated emission"). *E* is a waveguide which conveys the thus amplified microwaves to the detector. The entire process takes place in a vacuum. There is good reason to expect that this principle will make it possible to achieve very low noise factors.

from the oxide cathode can be demonstrated much more clearly by noise measurements than by measuring, say, the valve characteristic. Many other such examples might be enumerated.

Conclusion

To sum up we can say that there are three distinct groups of investigators concerned, each in their own way, with the study of noise.

The first group comprises the fundamental scientists; some, preceded by such famous men as Einstein, Smoluchowski, Zernike, Ornstein, Uhlenbeck and many others, are concerned with noise in the development of physical theory, while others use noise as a means for testing new theories experimentally or to throw light on unexplained phenomena. Until the processes taking place can be explained in physical terms it is not possible to

indicate the means of effectively combating the effects of noise.

The second group, comprising the valve manufacturers, attempts to apply the theoretical findings of the first group to the practical purpose of improving and modifying valve designs. In this way the manufacturers are enabled to supply data on the noise characteristics of their products.

The third group, that of the radio designers, puts to use the data provided by the valve manufacturers; this of course has long been the practice as regards other valve properties such as operating characteristics, capacitances, etc. Here, too, valve theory has always been left to the valve manufacturers. Armed with the valve data, the radio designer is

enabled to design his circuits so that they comply as well as possible with the requirements.

In many cases noise still represents a stumbling block to the radio designer. In radio broadcasting, radar, and some other fields of telecommunications the obstacle can to some extent be circumvented by making the signal stronger, that is to say by building more powerful transmitters, but of course there is always a limit to what can be done in this direction.

Nevertheless it may reasonably be expected that close cooperation between the above three groups, concerned in their different ways with the investigation of noise, will provide the radio listeners and television viewers of the future with a more and more perfect reception.

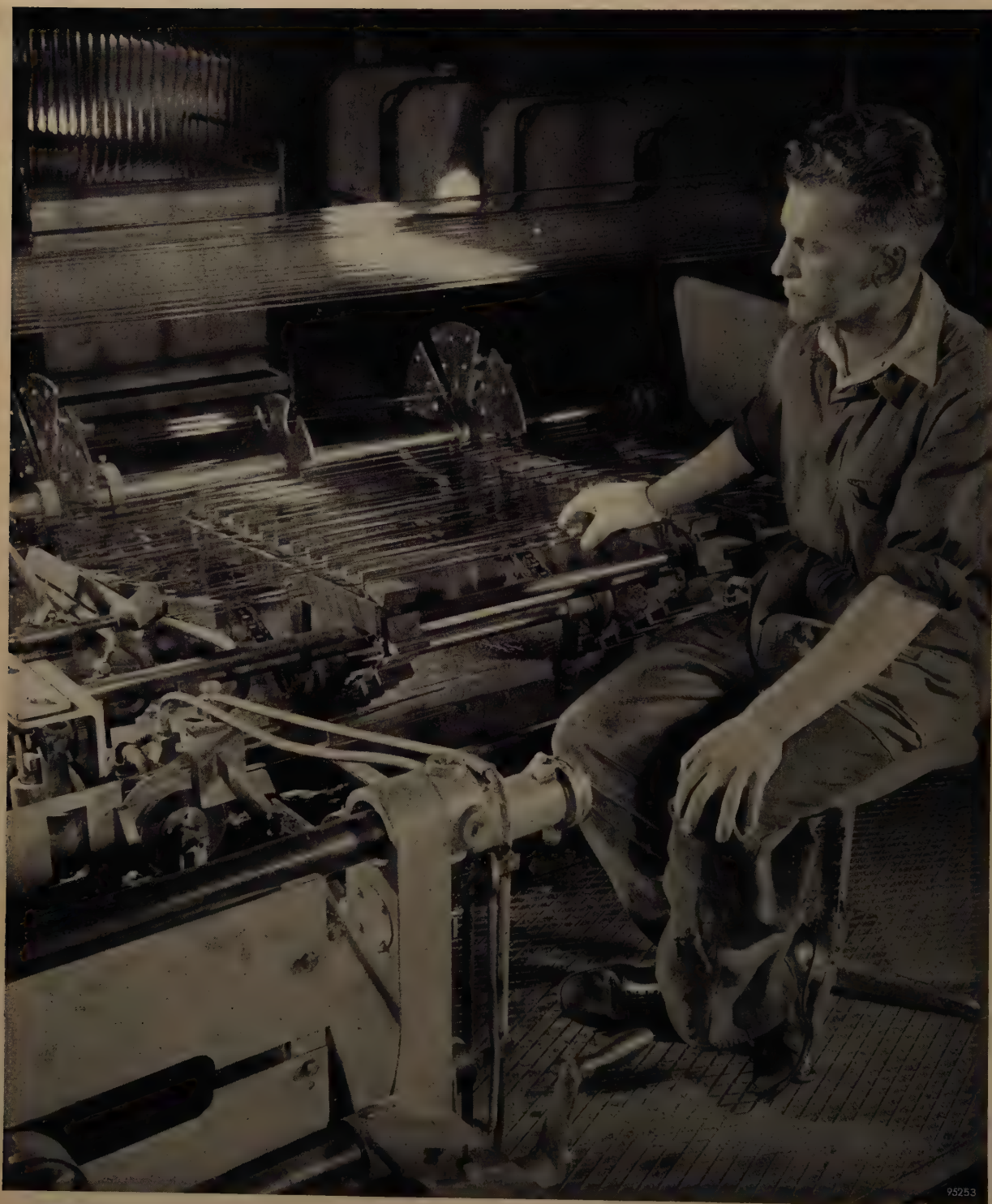
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Summary. The article substantially reproduces the address delivered by the author on the occasion of his inauguration as Professor of electronics at the Technische Hogeschool, Eindhoven. A survey is given of the phenomena, collectively termed "noise", which cause interference in e.g. radio and television reception. After touching briefly on the noise due to phenomena outside the receiver (man-made noise, atmospheric, cosmic noise), the author discusses at greater length the various forms

of noise arising inside the receiver, e.g. thermal noise, shot-effect, partition noise and secondary-emission noise, the noise appearing at very low frequencies and noise phenomena at very high frequencies (quantum noise and transit-time effects). He then discusses the influence of noise on the useful sensitivity of receivers, and concludes with a reference to the application of noise investigations in scientific research and industry.

AUTOMATIC SORTING OF GLASS TUBING



The machine shown here, in the Philips glass works at Roosendaal (Holland), is situated at the end of the glass tube drawing line. The glass tubing is fed from left to right (foreground) and automatically cut to a certain length. The moving belt transports these lengths towards the background in the photograph, where they are sorted according to diameter by rotating slotted discs. The tubing is used for the manufacture of fluorescent lamps.

THE LIFE OF BALLASTS FOR GAS-DISCHARGE LAMPS

I. TRANSFORMERS AND CHOKES

by T. HEHENKAMP.

621.327.032.43:621.314.2:621.318.42

Ballasts for gas-discharge lamps are among the articles that are required to give long service — e.g. twenty years — and to be low in price. This means that the designer must choose the insulating materials with care and ensure that they are operated at the highest temperature compatible with a reasonably long useful life. For these purposes life tests are necessary and, having regard to the long periods involved, the tests must be suitably accelerated. It is also highly important that the official regulations with which ballasts are required to comply should not hamper the development of cheaper apparatus of high quality by laying down unnecessarily low maximum temperatures.

A ballast generally consists of a choke or a transformer, often combined with a capacitor. Part I of this article is concerned with transformers and chokes. Capacitors will be dealt with in Part II, to be published later.

Factors that determine the life of a transformer

The life of a transformer (under which we include chokes for the purposes of this article) is taken to mean the time during which the transformer can operate before the insulation of the windings breaks down under the electrical field to which it is subjected. This time is determined by two quantities: the initial "reserve" of insulating strength and the rate at which this reserve falls to the zero value at which the insulation breaks down.

The reserve of the insulation depends on the *quality* of the insulating material used, on the *quantities* of these materials, and on the *manner in which they are processed*. The initial reserve can easily be ascertained by subjecting the insulation to voltages that are a multiple of those to which it will be subjected during operation. On the other hand, the rate at which the quality of the insulation deteriorates during operation — the aging rate — is much more difficult to ascertain. The following factors may be involved:

- 1) the electric field-strength in the insulation;
- 2) the mechanical load exerted on the insulating material;
- 3) climatic conditions;
- 4) the temperature of the insulating material.

In the case of transformers for gas-discharge lamps the first two factors are of subordinate importance, for these transformers are used at moderate voltages (except in neon installations, which are not considered here) and low power. Under these conditions ionization does not occur in the insulating material, and therefore the effect of the electrical field-strength on the aging rate is negligible. Because of the low power and the absence of moving parts, the mechanical forces acting on the insulation are small and

hence make hardly any contribution to the deterioration of the insulation during the greater part of the life of the transformer. Only when the insulation has undergone considerable structural deterioration is the breakdown accelerated by mechanical forces (we shall return to this subject later).

Climatic influences — apart from ambient temperature — affect transformers for gas-discharge lamps only in exceptional cases. Against moisture and mould it is the usual practice to mount the ballasts in closed boxes and, if necessary, to surround the ballast with a filler.

The temperature of the insulating material thus remains by far the most important factor determining the rate of deterioration. This is true not only of transformers for gas-discharge lamps but also of a large group of other transformers. It is therefore hardly surprising that a great deal of research has been carried out with the object of learning more about the influence of temperature on the rate of aging of insulating materials.

It was long believed that a critical temperature existed which set the boundary between a negligibly slow and a fast aging rate. In about 1930, however, Montsinger¹⁾ demonstrated that aging occurs at any temperature. His experiments showed that a temperature-dependence exists which can be represented by the equation

$$L = A e^{-BT}, \quad \dots \dots (1)$$

where L is the life, T the temperature of the insulation, and A and B are material constants. Represented graphically, with temperature on a linear scale and life on a logarithmic scale, this formula

¹⁾ V. M. Montsinger, Loading transformers by temperature, J. Amer. Inst. Electr. Engrs. **49**, 293-297, 1930.

yields a straight line (*fig. 1*). The constant B determines the angle which the line makes with the T axis. Montsinger found for B a value corresponding to a halving of the life for each 8°C rise in temperature.

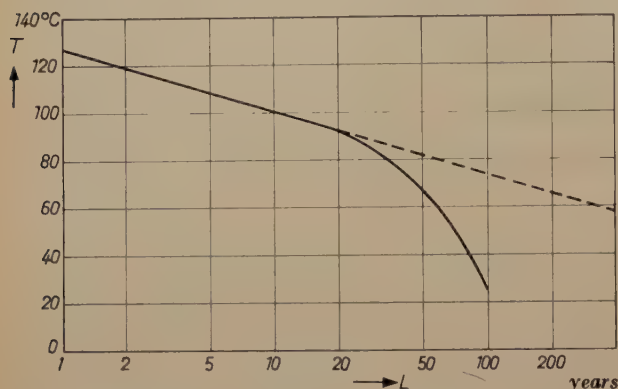


Fig. 1. Effect of the temperature T on the life L of organic insulating material in oil (graph taken from the article quoted under ¹⁾). The straight line corresponds to equation (1), the full line giving the results of tests, and the broken portion being an extrapolation to lower temperatures. Montsinger has suggested that the curve probably deviates from a straight line at low temperatures as shown for $L > 20$ years.

From later investigations ²⁾ it appeared that the rise in temperature, corresponding to a halving of the life, varies somewhat, approximately from 7 to 10°C . This variation is probably attributable in part to differences in the materials tested, but they are in any case also due to the fact that not all the tests were made in the same temperature range, the value of B being dependent to some extent on temperature.

On the basis of chemical considerations, Büssing ³⁾, and later Dakin ⁴⁾ arrived at a different formula, of the form

$$L = C e^{D/T}, \quad \dots \dots (2)$$

where C and D are again material constants and T is the absolute temperature of the insulation. Eq. (2) implies that the life curves plotted in a graph will be straight if L is plotted logarithmically and $1/T$ on a linear scale. In many cases this formula does in fact represent the results of life tests very satisfactorily, as illustrated by the example in *fig. 2*.

If this relation between temperature and life were valid in all cases, the investigation of the various materials and of the products manufactured with

them would be a simple matter, for a test at two temperatures would be sufficient to enable one to predict the life at any given temperature. Unfortunately, certain discrepancies occur, so that it is necessary to carry out life tests at a larger number of temperatures. This calls for much more extensive investigations. Another complication is that life tests on finished products are not always possible, for practical or economic reasons. One can therefore either examine the materials individually, or perform tests on simplified "models" of the products. We shall return to these alternatives later.

Choice of temperature range

Whichever method one adopts, it is very important to choose with care the temperature range in which tests are to be carried out.

It is tempting to choose a high temperature range, because of the quick results obtained in this way. The highest temperature at which tests are still possible is determined by the time needed by the product under investigation to reach this temperature. While the product is warming up, there will already be some aging of the insulation, but it is difficult to ascertain to what extent this takes place. For this reason the temperature chosen must be such that the life test is long compared with the time needed for the product to reach temperature equilibrium. In the case of small transformers this leads to a minimum time of several days.

The lowest test-temperature is decided by the accuracy one wishes to achieve in extrapolating the life curve to the normal working temperature. In view of the often wide spread in the measurements,

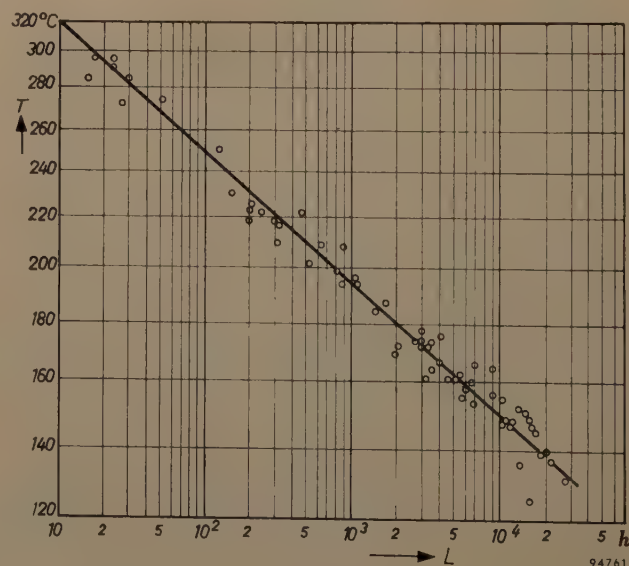


Fig. 2. Effect of the temperature T on the life L of impregnated asbestos paper (taken from article quoted under ¹¹⁾). The line corresponds to eq. (2).

²⁾ See e.g. G. L. Moses, *Westinghouse Engr.* **5**, 106, 1945.

³⁾ W. Büssing, *Beiträge zum Lebensdauergesetz elektrischer Maschinen*, *Arch. Elektrotechn.* **36**, 333-361, 1942.

⁴⁾ T. W. Dakin, *Electrical insulation deterioration treated as a chemical rate phenomenon*, *Trans. Amer. Inst. Electr. Engrs.* **67**, 113-122, 1948.

the lowest temperature should preferably not be much higher than the working temperature. Many transformers are required to give good service for some ten or twenty years. During this time they are usually not continuously under full load, but are loaded alternately or intermittently. The insulation, then, is subjected to the highest temperature only for a part of the time. For this reason a life of ten years under continuous full load is sufficient in most cases to ensure that, under practical conditions, the transformer will function efficiently for scores of years. In order to be able to extrapolate the results of accelerated life tests with reasonable accuracy to a time of ten years, the longest tests should be carried out at a temperature such that they last about two years.

To sum up, it can be said that the highest and the lowest temperature at which the tests are performed must be such as to correspond to lives of several days and several years respectively.

Life tests on component materials and on models

Among the various approaches to the problem of life tests are tests on the insulating materials themselves and tests on simplified or small-scale models of the transformers concerned. As will be shown below, however, this kind of testing does not generally lead to the required results.

Tests on insulating materials

It would offer considerable advantages if we were able to rely alone on life tests of the insulating materials themselves: the aging and loading of these materials to the breakdown limit requires much simpler equipment and is less expensive than the testing of complete transformers. Testing the material itself, however, is only of limited value, for two reasons. Firstly, there is the difficulty of ascertaining that property of the material which determines its useful life, and secondly it is often difficult to decide on the extent to which this property may deteriorate before the life test can be regarded as ended.

With insulating paper, for example, one can investigate in what way properties such as the breakdown voltage, tensile strength, bending strength, the dimensions (shrinkage) etc. change with time at various temperatures. According to Montsinger ¹⁾ the mechanical properties are decisive, whereas Stewart and Whitman ⁵⁾ maintain that the electrical properties are of primary importance (at least in the case of transformers cooled by air or, in general,

by a gas). Our own experiments indicate that the electrical properties are decisive if the insulation is loaded almost up to the breakdown limit, but that at the more common lower electrical loads (nominal voltage lower than about 1/3 of the breakdown voltage) the mechanical properties are of paramount importance.

If we know in a given case which property is the most important, there remains the problem of fixing the criterion for the end of the useful life. But even if this too is known, the difficulties are not yet over, for in practice the insulating material in transformers has so deteriorated towards the end of its life that it can no longer be handled. This greatly increases the experimental difficulties.

We shall illustrate these difficulties by some examples.

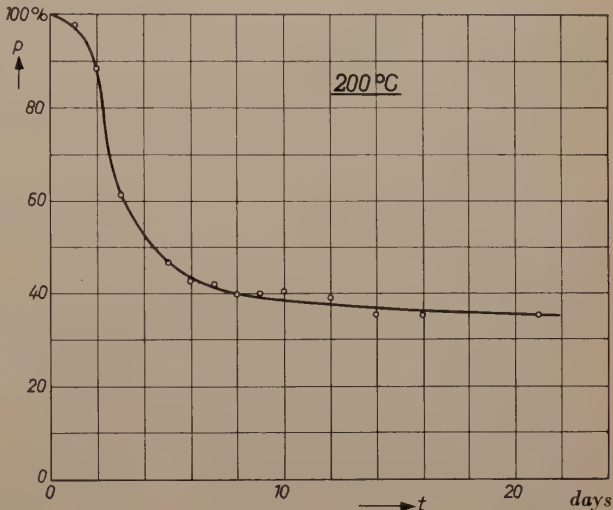


Fig. 3. Relative breakdown voltage p (alternating voltage, 50 c/s) of non-impregnated insulation cardboard 0.5 mm thick, as a function of the time t of exposure to air at 200 °C.

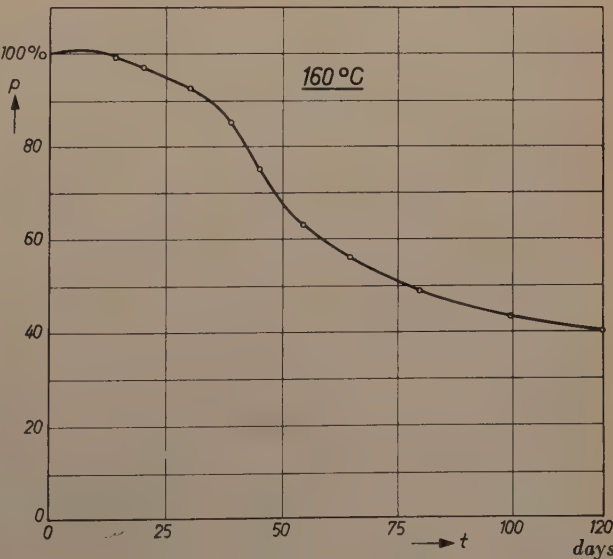


Fig. 4. As fig. 3, but in air at 160 °C.

⁵⁾ H. C. Stewart and L. C. Whitman, Aging characteristics of dry-type transformer insulation at high temperature, Trans. Amer. Inst. Electr. Engrs. **67**, 1600-1607, 1948.

The curve in *fig. 3* shows how the breakdown voltage of non-impregnated insulation cardboard depends upon the aging time, the test being in air at 200 °C. After about ten days the breakdown voltage has fallen to 40% of its initial value. The mechanical properties have so deteriorated by that time as to make further measurements difficult. Only with a number of precautions was it possible to extend the test to 20 days. Even after this time, however, the breakdown voltage still has a value which would be completely safe in the majority of transformers.

Fig. 4 shows the results of a similar test, but now at 160 °C. Here again it was found very difficult to continue the test when the breakdown voltage had fallen below 40% of its initial value.

In *fig. 5* the data from *fig. 3* and *fig. 4* are compared:

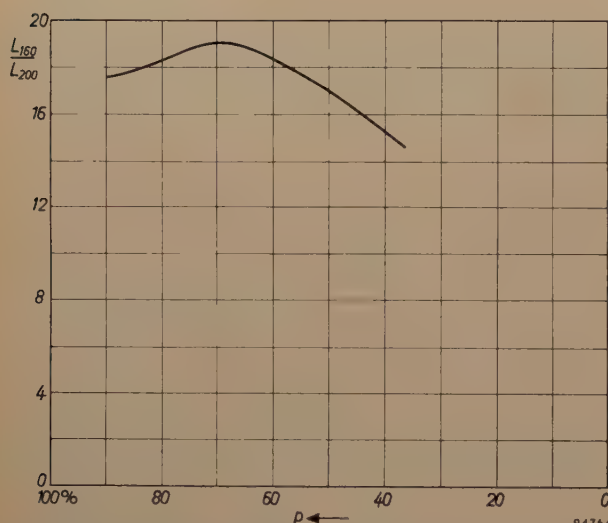


Fig. 5. The ratio of the times after which the breakdown voltage of the material in *figs. 4* and *3*, at 160 °C and 200 °C respectively, have fallen to the fraction p , as a function of p .

the time L_{160} in which, at 160 °C, the breakdown voltage falls to a fraction p of its initial value is divided by the time L_{200} needed for the same drop at 200 °C, and the ratio L_{160}/L_{200} is shown as a function of p . It can be seen that this ratio is by no means independent of p . This makes extrapolation of the curve to, say, $p = 10\%$, a hazardous matter.

As a second example we mention tests with "enamelled wire" (copper wire coated with an insulating lacquer). Loops of this wire were first examined for weak spots in the insulation by immersing them in a salt-water bath (*fig. 6*) and subjecting the insulation to 300 V alternating voltage. After those loops had been removed that were unable to withstand this load, the others were aged in an oven. At certain intervals they were removed from the oven in groups of ten, and subjected to a

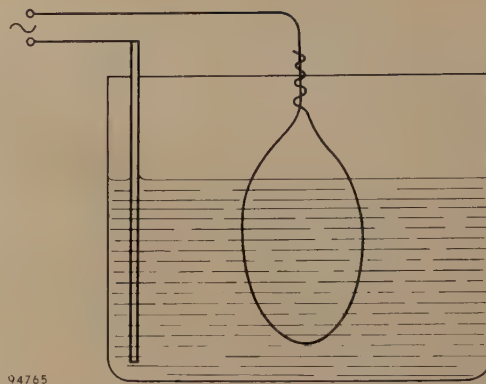


Fig. 6. Set-up to measure the breakdown voltage of the insulation on the copper wire to which *figs. 7, 8* and *9* refer. Wires, bent to form a loop, are immersed in a salt-water bath and loaded to breakdown by a slowly increasing alternating voltage.

breakdown-voltage test in the water bath. *Fig. 7* and *fig. 8* show the values of breakdown voltage found as functions of the aging time in air at 200 °C and 160 °C respectively. Here, too, the ratio of the times in which a certain drop in breakdown voltage occurs at each of these temperatures is not constant (*fig. 9*).

Our third example concerns the *weight* of the insulating paper. The loss of weight is an easily ascertainable criterion for the degree of deterioration, even when the material has already lost its structural integrity. *Fig. 10* shows the ratio of the times in which, at 160 and 200 °C, the weight decreases to a fraction p of its initial value, plotted as a function of p . The form of the resultant curve differs from that in both *fig. 5* and *fig. 9*.

The examples given and the results of many other tests have made it clear that it is necessary to know exactly what property of the material determines its useful life and at what load with respect to this property the material breaks down under practical conditions. To determine the slope of the life curve reliably (*fig. 2*), one must subject the material during the life test to the same load to which it would be subjected in actual operation. If we choose both the load and the permissible degree of breakdown in a relatively arbitrary manner, an entirely different slope may be found, and also the position of the curve with respect to the life axis may be quite wrong.

Having regard also to the considerable spread often found in measurements on materials, we see that it is extremely risky to draw from such tests general conclusions on the life curve of the finished product, and that the accuracy of such conclusions can often be the subject of dispute^{3) 6)}. These tests

⁶⁾ J. J. Smith and J. A. Scott, Temperature aging characteristics of class A insulation, *Trans. Amer. Inst. Electr. Engrs.* 58, 435-444, 1939.

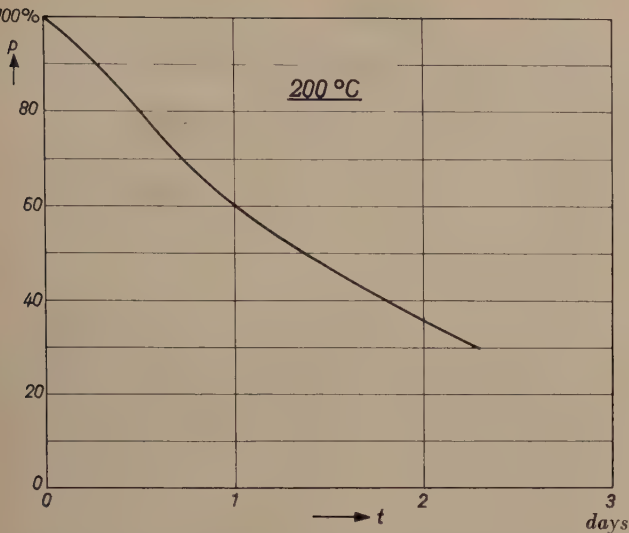


Fig. 7. Relative breakdown voltage p (A.C., 50 c/s) of 0.4 mm thick copper wire insulated with lacquer, as a function of the time t of exposure to air at 200 °C.

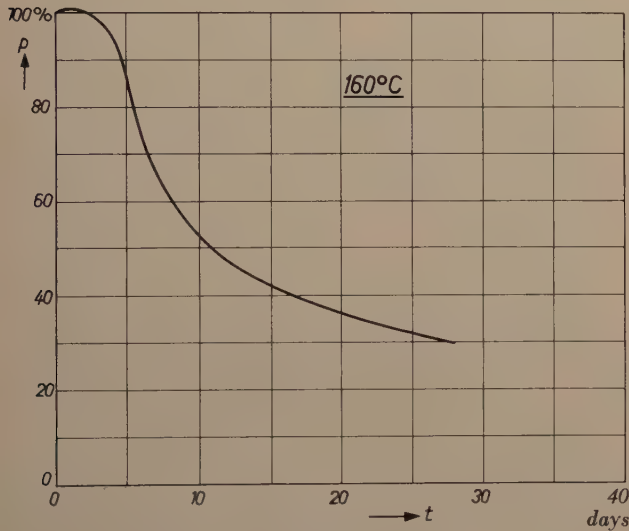


Fig. 8. As fig. 7, but in air at 160 °C.

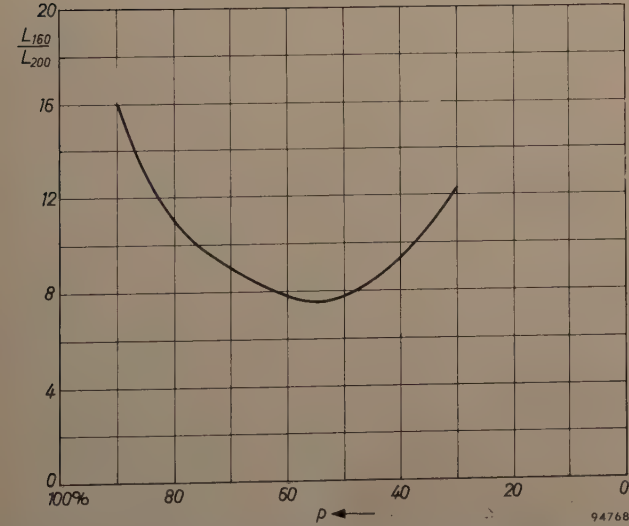


Fig. 9. The ratio of the times after which the breakdown voltage of the copper wires in figs. 8 and 7, at 160 °C and 200 °C respectively, have fallen to the fraction p , as a function of p .

can, however, give a rough impression of the performance that may reasonably be expected from the tested material when incorporated in the transformers. The final appraisal of the serviceability of the product, however, calls for more effective tests.

Model tests

Since, as we have seen, tests on individual insulating materials do not yield adequate results, while tests on complete transformers are often too costly, the solution sometimes adopted is to carry out the tests on a cheaper (often smaller) “model” of the transformer. The processing of the insulating material used and its loading in the model must correspond as closely as possible to the situation in the actual transformer. As regards loading, this can cause difficulties, since the conditions in models will generally be different from those in the actual transformer. The electrical load can usually be imitated quite well, but the mechanical load not so easily: the forces acting on the windings as a result of electric currents and electric fields may have quite different magnitudes in the model. One method of achieving a better approach to the conditions of the actual transformer is to cause the model to vibrate

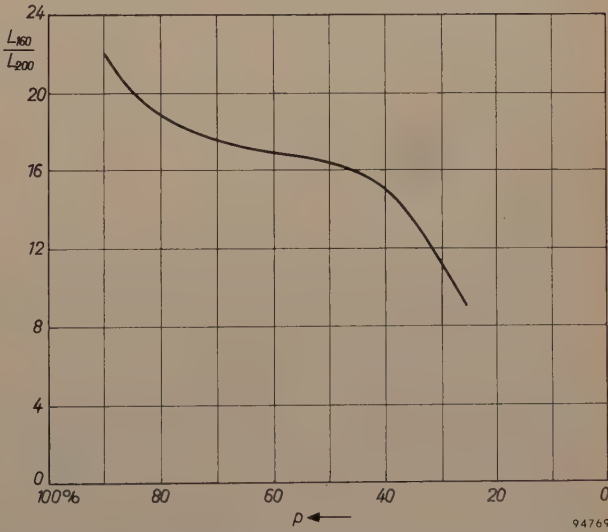


Fig. 10. The ratio of the times after which the weight of the insulating paper, at 160 °C and 200 °C respectively, have diminished to the fraction p , as a function of p .

with the appropriate amplitude and frequency⁷⁾. Another method is to construct only the coils and just for the purpose of testing to make them into complete transformers by placing them over a core⁸⁾. Without measures of this nature, the results

7) R. E. Whipple, Aging characteristics of electrical insulation, N.R.L. Report 3708, July 1950.
8) J. A. Scott and B. H. Thompson, Temperature-aging tests on class-A-insulated fractional-horsepower motor stators, Trans. Amer. Inst. Electr. Engrs. 61, 499-501, 1942.

of the tests can be very misleading, as will be illustrated by the following series of experiments.

Large numbers of test models were made to simulate chokes for fluorescent lamps; their construction is illustrated in *fig. 11*. The first four

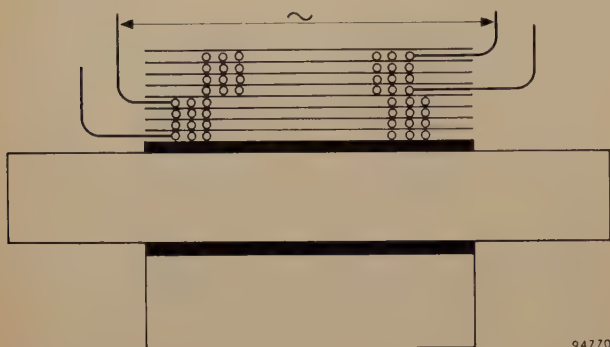


Fig. 11. Cross-section of coils for model tests. On a hollow cylindrical former of insulating material two sets of four layers of "enamelled" copper wire are wound, with intervening layers of insulating paper. The insulation between the fourth and the fifth layer was tested for breakdown. A straight iron core was inserted in the former to prevent deformation.

layers of turns are electrically insulated from the next four layers. The insulation between these groups of layers consists of lacquer + insulating paper + lacquer. The breakdown voltage of this insulation can accordingly be determined without current flowing through the windings, thus avoiding the forces which the current would otherwise occasion. The coils were supported by the core which incorporates a mounting strip (*fig. 12*), and treated

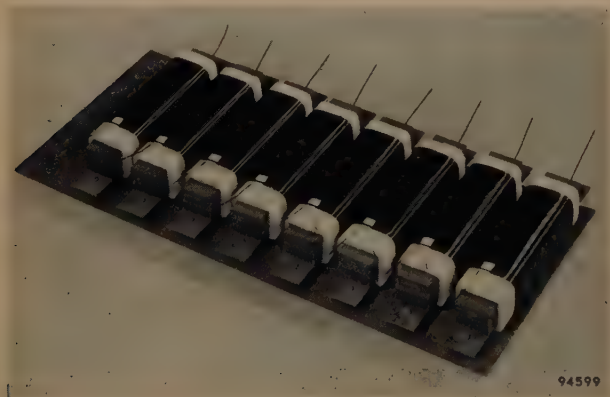


Fig. 12. Choke "models" which were life-tested under conditions of electrical loading only.

with extreme care. Thus, as opposed to the situation with the real chokes in ballasts, the loading of the models was almost exclusively electrical.

The results of the breakdown tests after various aging periods at 200 °C are shown in *fig. 13*. Three regions can be distinguished: in the first region the breakdown voltage drops very rapidly and there is a

wide spread in the measurements; in the second region the breakdown voltage is almost constant and the spread small, and in the third region there is again a drop in breakdown voltage and a wider spread.

In the first region the insulation deteriorates not only electrically but also in its mechanical properties and in thickness, but it still remains intact. At this stage, then, we can speak of breakdown in the insulating material itself (*fig. 14*).

With continued aging the paper insulation begins to crack, which can give rise to paths for direct flash-over from one winding to another. The breakdown voltage is then determined largely by the distance to be bridged, hence by the thickness of the insulation beside a crack. This thickness decreases only very slowly with time, so that there is very

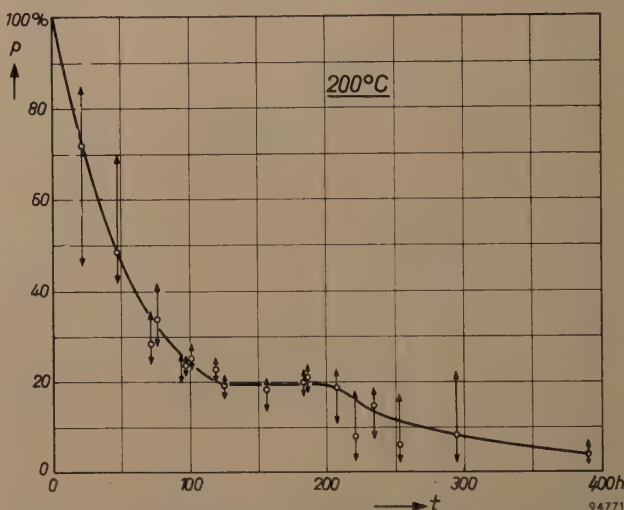


Fig. 13. Relative breakdown voltage p of the insulation between two layers, as a function of the aging time t in air at 200 °C.

little falling-off in the breakdown voltage for some considerable time (the "second region" in *fig. 13*). As the deterioration continues, the number of cracks shows a marked increase, the insulation disintegrates into ever smaller parts and is finally reduced to shreds. In this third region — as opposed to the others — mechanical forces have a considerable effect on the insulation, for vibrations and shocks accelerate the disintegration process. The time during which the tests can be continued is therefore partly dependent on the care exercised when handling the chokes. The sharp drop in the relative value of the breakdown voltage at 200 hours is presumed to be due to mechanical damage caused when removing the chokes from the oven for testing.

Corresponding tests at 160 °C yielded results as shown in *fig. 15*. Here, too, the first two stages can be distinguished: the third had not yet been reached after 200 days. If, as in *figs. 5, 9 and 10*, we again

plot as a function of p the ratio of the times in which at 160 and 200 °C, respectively, the breakdown voltage drops to a fraction p , we obtain a curve as shown in *fig. 16*. Here too, instead of a straight horizontal line we have a curve that allows no more than a rough extrapolation to a small value of p . Thus, tests on models also involve the difficulty of

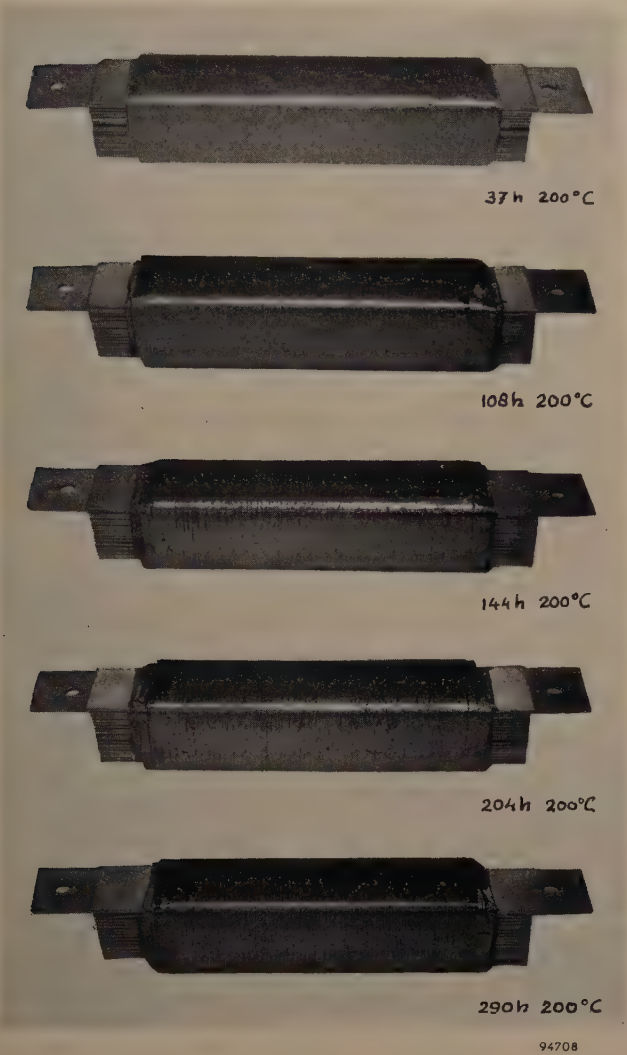


Fig. 14. Choke “models” as in *fig. 12* after aging at 200 °C for 37, 108, 144, 204 and 290 hours respectively. The four outside layers have been removed to show the insulation between the fourth and fifth layer, which was that electrically tested. As aging continues, the number of cracks increases and finally the insulation disintegrates.

accurately imitating the conditions of practical operation. In order to compare different insulating systems by means of tests on models it is therefore not enough to carry out all tests under the same conditions, but the conditions themselves must be the right ones ⁹⁾.

⁹⁾ L. J. Berberich and T. W. Dakin, *Insulation*, March 1956, p. 21.

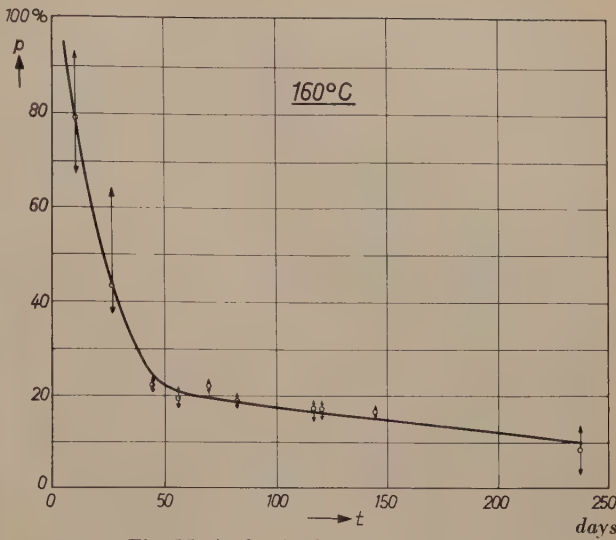


Fig. 15. As *fig. 13*, but in air at 160 °C.

Life tests on complete products

The foregoing makes it obvious that, if it is at all economically justified, the life tests should be carried out on complete products. The economic objections are not serious if only small transformers are concerned, such as those of ballasts for gas-discharge lamps.

To make the temperature of the insulation higher than normal, one can either increase the heat generated in the transformer itself, or apply heat externally by placing the transformer in an oven. In the following we shall give an example of both methods. In the first the heat generated in the transformer is increased by raising the current density in the windings (the test chokes were wound with thinner wire for this purpose). The second example refers to chokes that were left, unloaded, in an oven and switched on only for a short time once a day.

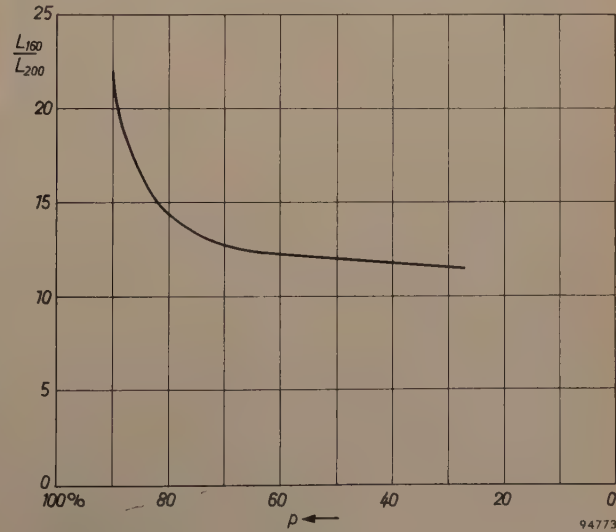


Fig. 16. The ratio of the times after which the breakdown voltage of the insulations in *figs. 15* and *13*, at 160 °C and 200 °C respectively, have fallen to the fraction p , as a function of p .

Other methods of increasing the current density are:

- 1) To raise the supply voltage. The drawbacks of this are that, owing to the strong magnetic saturation of the core, the current becomes very dependent on voltage fluctuations, and that the mechanical forces become abnormally large.
- 2) To introduce air-gaps in the core, or to enlarge the air-gaps. In this case, also, the forces acting on the insulation will not be of the correct magnitude.

These drawbacks are avoided by heating the chokes in an oven. In the design of conventional ovens, however, allowance is not generally made for any heat developed in the object itself, and this heat can so adversely affect the internal temperature distribution as to make the temperature control inaccurate. These difficulties can be overcome by not loading the transformers while they are in the oven (or loaded only for short times, with long intervals in between).

Tests at excess current densities

In order to study the behaviour of the insulating material used at the time in Philips ballasts for fluorescent lamps, forty chokes were constructed in 1949 which differed in only two respects from the normal types used for 40 W, 220 V (50 c/s) fluorescent lamps. In the first place the impregnation with asphalt compound was omitted in order later to be able to investigate separately the influence of different methods of impregnation. In the second place the forty chokes were divided into five groups, each being wound with wire of a different thickness. As a result of the five current densities so obtained, the insulation, which normally reached about 90 °C, now reached temperatures from 100 to 270 °C, thereby giving the desired accelerated aging. Owing to the small diameter of the coils there was no internal temperature-gradient worth mentioning. The temperature of the insulation was ascertained from the increase in the resistance of the winding and from the ambient temperature.

All 40 chokes were connected to 220 V (50 c/s), i.e. to the normal loading voltage when the fluorescent lamp is still short-circuited by its starter. At irregular intervals the installation was switched on and off in order to reproduce the switching transients occurring in actual operation.

The results of these tests are collected in *fig. 17*. It can be seen that they are in good agreement with the life formula (2). After 28 months the tests had to be ended owing to a defect in the test equipment which damaged the 8 chokes that were still under test. The measurements cover lifetimes in the ratio 1 : 400; extrapolation to 10 years is therefore readily possible.

According to *fig. 17* the life of the choke at 160 °C is only about 6 times as long as at 200 °C, a fact which had not emerged from any of the tests on individual materials or on models. This again underlines the limited value of the latter tests.

Tests on unloaded chokes in ovens

In 1956 the tests described with reference to *fig. 17* were recommenced. Large numbers of chokes were constructed — again chokes for 40 W, 220 V (50 c/s) fluorescent lamps — but of the design as

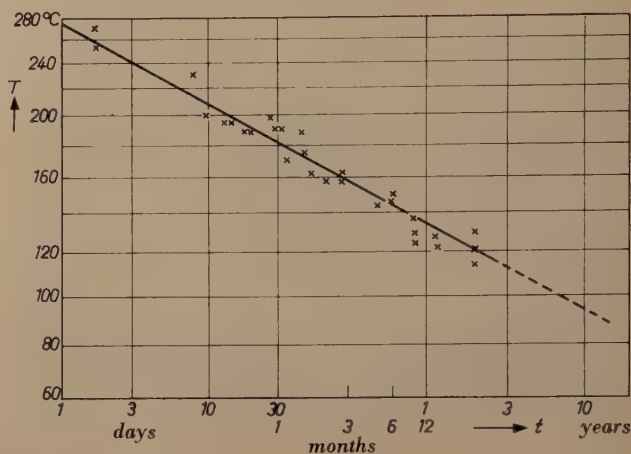


Fig. 17. The life L of non-impregnated chokes for 40 W 220 V (50 c/s) fluorescent lamps as a function of the insulation temperature T . The various temperatures were obtained by running the chokes at different current densities.

used nowadays. They were cast in the appropriate boxes with a filling material based on a polyester resin¹⁰⁾ and so made into a finished product.

The insulation was raised to the testing temperatures by placing the chokes unloaded, in groups of ten, in ovens. Each group was daily connected for one to two minutes to 220 V (50 c/s) by means of a rapidly switching contact. The closing of this contact produces initial current surges about four times as high as the normal operating current; when the contact opens, voltage surges appear which are about seven times as high as the operating voltage. These loads so far exceed the severest conditions encountered in practice, both in magnitude and in duration, that in our opinion the absence of the much lower operating voltage while in the oven has scarcely any influence.

The results hitherto obtained yield points lying on a straight line (*fig. 18*) which runs parallel to the life curve of *fig. 17*. Owing to the better insulating materials and to the polyester-resin filling, the life of the choke at each temperature has increased more than eight-fold.

Life under practical conditions

In *figs. 17* and *18* the life of the choke is the time at the end of which half of the original number of coils is still intact. Extrapolation to the temperature

¹⁰⁾ T. Hehenkamp, Philips tech. Rev. 18, 280, 1956/57.

commonly occurring in practice thus provides the time at which, at that temperature, an average of 50% of the chokes will be defective. To the user, however, this information is not so interesting; he is more interested in the time during which he can

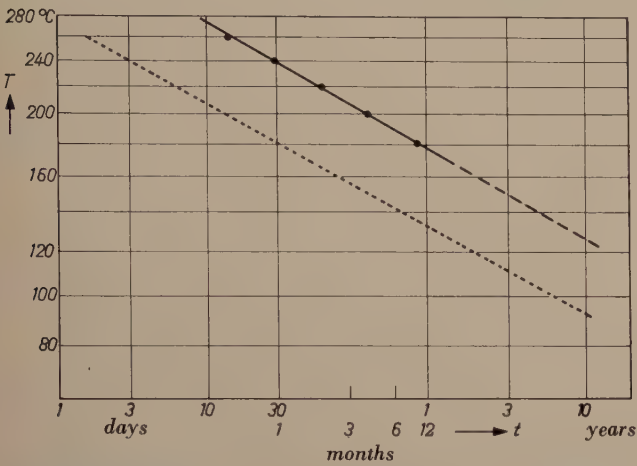


Fig. 18. The life L of chokes for 40 W, 220 V (50 c/s) fluorescent lamps (boxed, with polyester-base filler) as a function of the temperature T of the insulation. The various temperatures were obtained by heating in ovens. The life curve from fig. 17 is shown for comparison (broken line).

expect no serious faults to occur in his installation. To ascertain this time one must know the spread in the life of the chokes, i.e. the reject curve. Although some predictions can be made from the results of the tests described, reliable figures can only be obtained after investigating much larger numbers.

This is done at Philips by regularly subjecting samples from the ballasts manufactured for fluorescent lamps, sodium lamps and HPL lamps (high-pressure mercury-vapour lamps with fluorescent outer envelopes) to a life test. As long as the design is not radically modified, it is sufficient to perform this test at only one temperature. This was fixed some years ago at 180 °C.

During the years 1953 to 1956 almost 1000 of such sample tests were carried out. They showed that in 30 days — according to fig. 17 the time in which 50% failures occur at 180 °C — about 3% of the ballasts became defective. It can be concluded from this that the life curve in fig. 17 denotes the times after which about 3% of the ballasts in an installation will become defective. With such a chance of failure an installation gradually becomes due for replacement.

The corresponding figures are not yet accurately known for the ballasts with polyester filling which were recently put into production. It is expected that their life will be at least twice as long.

From these data the permissible temperature of the insulating material can be determined for any installation. For this purpose it is only necessary to

know how many years the installation must operate reliably and how many hours it will be in operation during that time. An ordinary lighting installation, for example, will be required to operate for an average of 8 hours per day for 20 years. Since the temperature of the insulation does not reach its final value until about two hours after switching on, these first two hours need hardly be taken into account. As far as aging is concerned, therefore, one may reckon with an average of six hours per day, giving a total aging time of $\frac{6}{24} \times 20 = 5$ years continuous operation.

Ballasts as made in recent years have a life of 5 years continuous operation when the temperature of the insulation is 105 °C. At an ambient temperature of 35 °C this means, therefore, a rise of 70 °C in the temperature of the insulation. When the measurement is performed (as is the usual practice) at a supply voltage 10% higher than the normal mains voltage, the temperature in this measurement may without harm rise to about 80 to 90 °C; most official specifications, however, unnecessarily prescribe a temperature rise of only 70 °C.

In special cases a higher temperature may be permissible, or a lower temperature necessary. The first case arises when a much shorter life is acceptable, for example with show-window lighting which, for advertising reasons, will in many cases be replaced after a few years. On the other hand the temperature of the insulation must remain below a lower maximum value if the lighting is kept on for a longer average time per 24 hours than is assumed above, the same life being required of the installation.

The same conditions apply to the latest ballasts with polyester filling, except that the temperature of the insulation can always be 10 °C higher (i.e. 90 to 100 °C above the ambient temperature at 10% over-voltage).

Classification of insulating materials

For many years now it has been the practice in national and international specifications to classify the various insulating materials, and to lay down a maximum permissible temperature for each class.

It will be evident from the foregoing that a classification of insulating materials into, say, four or six categories, each with a maximum permissible temperature, can be little more than arbitrary. After all, there are many possibilities of appreciably improving the insulation of a transformer, thereby prolonging its life, without the necessity of using a material of a higher category. Some examples will make this clear.

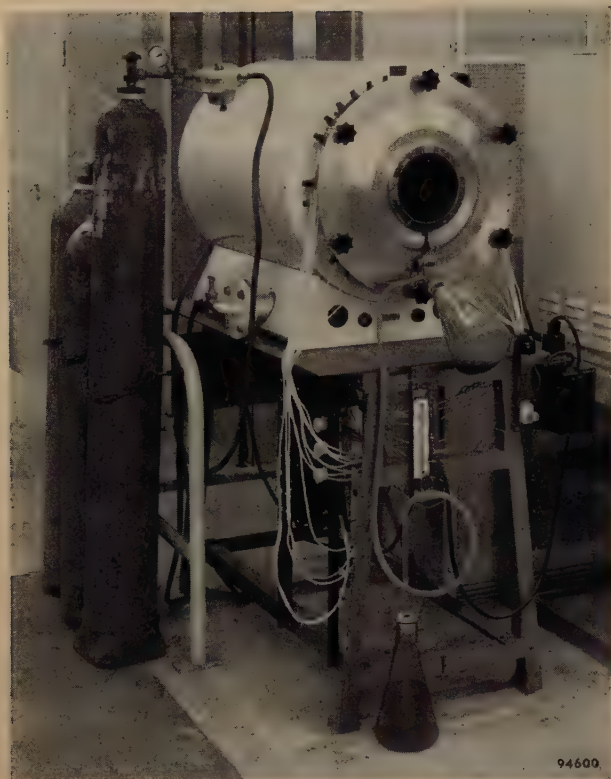


Fig. 19. Oven for life-testing transformers in an oxygen-free atmosphere.

In ballasts for gas-discharge lamps frequent use is made of copper wire insulated by a coating of lacquer ("enamelled" wire). This wire comes into a certain insulation category, but the quality of the lacquer coating varies considerably in practice, giving rise to differences in life. There are also synthetic wire-insulating materials of higher quality than good oil-based lacquers. Some specifications take these into account by permitting a somewhat higher temperature for a synthetic insulating material. This is not without its dangers, however, for some kinds are not even as good as oil-based lacquers of mediocre quality.

Another factor of influence can be the filling material which may be used to seal off the coils. It is known that the processes of deterioration are not so rapid if there is no exposure to oxygen ¹¹⁾.

¹¹⁾ H. C. Stewart, L. C. Whitman and A. L. Scheideler, Aging evaluation of dry-type transformer insulating systems, Trans. Amer. Inst. Electr. Engrs. **72-III**, 267-276, 1953.

Exclusion of air by means of a filling material will therefore increase the life of the insulation, provided of course that the filler does not itself take part in any reaction which offsets the favourable effect. Whether the latter is the case can be ascertained by testing the equipment in an oxygen-free atmosphere with and without filler; an oven used for this purpose is shown in *fig. 19*. Such tests show that unsuitable filling materials shorten the life of the equipment.

In view of the fact that new insulating materials regularly appear on the market, opening up new possibilities, it is plain that classification must go very far indeed if the designer is not to be needlessly hampered in developing an optimum product.

For products that are small and cheap enough to justify life tests the classification should therefore be taken as a rough guide, after which the really permissible temperature should be determined by means of accelerated life tests. A recommendation to this effect has already been made by Stewart ¹¹⁾. In the latest German specifications for ballasts ¹²⁾ this procedure has in fact been adopted ¹³⁾.

For larger and more costly transformers it will probably be possible to obtain satisfactory results by tests on models, although the necessary caution will have to be observed in view of their limited reliability.

¹²⁾ VDE 0712, Part 2, April 1957.

¹³⁾ C. H. Sturm, Zur Neuausgabe von VDE 0712 „Zubehör für Leuchtstofflampen“, Elektrotechn. Z. **A78**, 363, 1957 (No. 10).

Summary. The life of a transformer (or choke) is the time the transformer can operate before the insulation breaks down. This time is determined by the "reserve" of insulating strength initially present, and by the rate at which this reserve decreases. The reserve depends on the quality, quantity and processing of the insulating materials employed. In the case of transformers and chokes for the ballasts of gas-discharge lamps, the rate of aging is determined primarily by the temperature of the insulation. Life tests can be accelerated by performing them at higher temperatures, e.g. by running at excess current densities or placing the product in an oven. The range of temperatures for such tests should be such that the shortest life amounts to a few days and the longest to a few years. Tests on the insulating materials themselves or on models have only limited value; for this reason, if at all economically justified, the tests should be carried out on the complete product.

LATTICE IMPERFECTIONS IN CRYSTALS, STUDIED ON ALKALI HALIDES

by Y. HAVEN.

548.7: 537.311.3

Many properties of solids, including such technically important ones such as electrical conductivity and dielectric losses, cannot be explained on the basis of a purely periodic structure of the crystal lattice. The observed phenomena have therefore come to be attributed to certain deviations from exact periodicity — that is to say, to lattice imperfections. The effects of lattice imperfections on the behaviour of solids can very conveniently be studied on crystals of the alkali halides, since these substances can easily be obtained in a pure state and since in their case the phenomena are often less complicated than in other cases. In this article some results of investigations on these crystals are given, with particular reference to the dielectric properties of an alkali halide (NaCl) in which certain lattice imperfections can be produced by introducing an alkaline-earth halide (CaCl₂).

Lattice imperfections in crystals

Since the discovery by Von Laue, Friedrich and Knipping (1912) of X-ray diffraction in crystals and the investigation of this phenomenon by Von Laue, Bragg and many others, a great deal of work has been carried out on the spatial arrangement of the atoms in crystalline solids. It was found that the atoms in the crystal (for convenience we shall often speak of atoms even though ions are concerned) form a three-dimensional network or "lattice" and that the atoms are arranged periodically in the lattice. This is entirely in agreement with the conjecture of Bravais some fifty years earlier concerning the structure of crystals.

It subsequently appeared, however, that many phenomena exhibited by solids could not be explained on the basis of a purely periodic structure of the crystal lattice. Frequently these phenomena are closely related to those properties that are put to use for various technical purposes, especially in electrical engineering and the electronics industry. These are the properties that determine the electrical behaviour of the solid, such as electrical conductivity and dielectric losses. But there are other properties, too, such as mechanical strength, light-absorption, luminescence and certain details of crystal growth, that cannot be quantitatively explained on the assumption of a purely periodic structure. Many of the observed properties of crystals have therefore come to be attributed to certain deviations from periodicity, collectively known as lattice imperfections. Some considerations on the role of lattice imperfections in solid state phenomena were given in an earlier article published in this journal ¹⁾.

The deviations from periodicity, whose existence was initially postulated purely hypothetically, are

of many kinds. We shall discuss some of them briefly following the general lines of a survey given by Seitz ²⁾.

Vacancies. This term is used to designate an empty lattice site, that is, the absence of an atom at a position where, according to the crystal periodicity, there should be an atom.

Interstitial atoms. An atom is referred to as an interstitialcy when it occupies a site lying between the normal lattice sites of the crystal.

Foreign atoms. Foreign substances can be added as "impurities" to a crystal of a given compound. If the substances are distributed atomically in the crystal (thus not locally segregated) the effect will be that atoms are present in the crystal lattice which are foreign to it. These foreign atoms may occupy normal lattice sites (by substitution) or they may be in interstitial sites.

Vacancies, interstitialcies and foreign atoms have an important bearing on the phenomena to be discussed in this article.

Free electrons, electron holes, colour centres, excitons. If an electron is added to a (neutral) crystal lattice, a positive ion of the lattice is entirely or partly neutralized. In general, however, the position of the electron is not fixed, but can change from one ion to another. This, too, can be regarded as a (mobile) lattice imperfection.

Such an added electron may, for example, originate from a foreign atom ("donor") which gives up an electron to the lattice. There are also foreign atoms that deprive the lattice of an electron ("acceptors") This leaves behind a "hole" in the electron distribution, which means to say that one of the ions is then short of an electron. This state, too, provided the temperature is high enough, can shift

¹⁾ G. W. Rathenau, Imperfections in matter, Philips tech. Rev. 15, 105-113, 1953/54.

²⁾ F. Seitz, contribution in the book: Imperfections in nearly perfect crystals, Wiley, New York 1952, p. 3.

from ion to ion throughout the lattice. At low temperatures, however, one must rather think of the electron hole as localized; in KCl at -200°C , for example, the hole is bound to two neighbouring Cl ions, giving rise to a Cl_2^- ion³⁾.

An electron and an electron hole can also be simultaneously produced by an atom of the original lattice. The pair can then be regarded as a non-localized ionized state.

When a crystal contains yet other lattice imperfections, a certain interaction will arise between the electron and one of these imperfections, e.g. a vacancy. Sometimes the electron may then be trapped. Such an electron, owing to the selective light absorption it causes, is called a *colour centre*.

There also exist excited states of the lattice, in which one of the ions is at a higher energy level, but in which the excess energy is smaller than the energy needed for the formation of a free electron plus an electron hole. If this disturbance is free to wander throughout the lattice it is called an *exciton*.

Some other lattice imperfections are the following.

Grain boundaries. A polycrystalline solid is built up of more or less distinct crystals separated by what are known as "grain boundaries". At these boundaries there is an abrupt change in the orientation of the crystal axes, so that the periodicity is interrupted at these positions, quite apart from the degree of perfection of the crystal grains themselves.

The smaller the crystals the more numerous are the disturbances due to grain boundaries. In the extreme case the crystals are so small that the volume occupied by the affected areas is of the same order of magnitude or greater than that of the unaffected areas. The substance is then called amorphous. A familiar example is glass⁴⁾.

Dislocations. Dislocations are linear lattice defects. A simple form of dislocation can be visualized by thinking of a lattice plane as pushed some way into the crystal, between the other lattice planes (like a snippet of paper inserted between the pages of a book). Near the edge of this interposed lattice plane the crystal is severely deformed. If foreign atoms are present in the crystal, they show a preference to settle at such regions of marked deformation.

Dislocations go a long way towards explaining many properties of metals⁵⁾. In non-metallic substances, too, the occurrence of dislocations must also be borne in mind.

Phonons. Finally, lattice imperfections can also be caused by sound waves passing through a crystal. The deformation thereby caused moves progressively through the lattice at

the speed of sound. The sound energy can be thought of as concentrated in a relatively small region (wave packet) which is termed a "phonon" (a term analogous to "photon", a packet of electromagnetic energy).

The thermal agitation of the atoms also represents a disturbance of the lattice. This may be regarded as a swarm of phonons moving through the crystal like the molecules of a gas.

It need hardly be said that, if several of these lattice imperfections are combined, they may give rise to complicated phenomena not easy to disentangle. For this reason the aim in studying lattice imperfections and their effect on various properties is to confine oneself as far as possible to cases in which it may be assumed that only one particular type of imperfection is predominant.

Such cases can often be produced artificially by taking a very pure material and subjecting it to a treatment designed to give rise to a specific imperfection. This treatment may, for example, consist in incorporating as an impurity a small quantity of another pure substance. Several investigators have adopted this method successfully with the alkali halides, which in very many respects lend themselves particularly well to investigations of this kind. Not only can these compounds be prepared in a pure state by relatively simple means, but they also have the advantage of possessing a simple crystal structure (monovalent ions). Since, moreover, the ionic character of these compounds is very pronounced, it is possible, in view of the fact that the attractive forces between the ions follow Coulomb's law, to compute the interaction between the particles without involving excessively complicated mathematics.

There are certain other advantages attached to the use of the alkali halides. In the first place they are transparent, which makes it easy, for example, to observe their spectral transmission. In coloured crystals, for example, the position of the absorption bands in the spectrum can be determined. In some cases particular imperfections, such as dislocations, can be made visible under the microscope by such artifices as the introduction of metal that nucleates at the lattice imperfections⁶⁾. A further advantage of alkali halides is that various heat treatments, required in the investigations, can be carried out at relatively low temperatures.

Admittedly, information is obtained in this way on only one category of compounds and on relatively few kinds of lattice imperfections, but if detailed knowledge is thereby obtained this can serve as a point of departure for understanding, at least quali-

³⁾ T. G. Castner and W. Känzig, *Phys. Chem. Solids* **3**, 178, 1957 (No. 3-4).

⁴⁾ It should be mentioned in passing that the structure of glass can be described in another way, i.e. according to the now generally accepted theory of Si-O networks (Zachariasen); see e.g. J. M. Stevels, *Philips tech. Rev.* **13**, 293, 1951/52.

⁵⁾ H. G. van Bueren, *Lattice imperfections and plastic deformation in metals*, *Philips tech. Rev.* **15**, 246-257 and 286-295, 1953/54.

⁶⁾ S. Amelinckx, *Phil. Mag.* (8) **1**, 269, 1956.

tatively, the part played by lattice imperfections in more complicated compounds.

In the following we shall discuss some investigations on alkali halides concerned with the relation between certain lattice imperfections and electrical properties, in particular dielectric losses.

Crystal structure and lattice imperfections in alkali halides

We shall first deal at some length with several kinds of lattice imperfections postulated in alkali halides, and indicate the reasons for concluding that these imperfections exist.

The alkali halides crystallize in the cubic system, and mostly have the so-called sodium chloride structure ⁷⁾. Each alkali ion is surrounded in the ideal lattice by six halogen ions, and each halogen ion by six alkali ions.

An ideal alkali halide crystal composed of ions is theoretically a perfect insulator. In reality, however, conduction, viz. ionic conduction, is found to occur, particularly at high temperatures. This can only mean that numbers of ions depart from their prescribed positions in the lattice. One can imagine that an ion occupies an interstitial lattice site and changes its position in the crystal as an interstitial ion. However, if an ion moves to an interstitial site, it must leave behind an unoccupied site or vacancy. Frenkel remarked as early as 1926 that such vacancies must also give rise to ionic conduction, for a neighbouring ion can occupy the vacancy and in its turn leave an unoccupied site behind. The vacancy then moves through the lattice.

This kind of lattice imperfection (called a Frenkel defect) obeys the rule that electric neutrality of the crystal must always be maintained, for in a region which contains interstitial ions together with the vacancies associated with them, there is no change in the nett electrical charge.

It cannot be said *a priori* which of the two types of ion form the interstitial atoms. Fundamentally there are two possibilities:

- 1) positive ion on interstitial site + positive-ion vacancy.
- 2) negative ion on interstitial site + negative-ion vacancy.

Another combination of lattice imperfections is conceivable which also preserves electric neutrality. One can imagine that a positive ion in a certain site in the lattice is removed from the crystal and a negative ion from another site is removed from the

crystal. This results in vacancies, neutrality being preserved without it being necessary to assume the presence of interstitialcies. The possibility of this kind of imperfection was first put forward by Schottky (1930).

One can also imagine a situation in which somewhere in the lattice a positive interstitial ion comes into the neighbourhood of a negative interstitial ion.

These considerations give two new possibilities:

- 3) positive-ion vacancy + negative-ion vacancy (Schottky defect),
- 4) positive interstitial ion + negative interstitial ion.

There are thus four possibilities to be taken into account in the explanation of ionic conduction in pure alkaline halides. It need not be assumed, however, that these four imperfections will be present at the same time. It has been found, for example, that many aspects of this conduction can be accounted for by assuming that only Schottky defects occur in pure alkaline halides.

Other forms of imperfection are possible when, as already mentioned, an impurity is artificially introduced, e.g. when a small quantity of a divalent metal is incorporated in the monovalent alkaline halide. If, for example, a small quantity of CaCl_2 is added to NaCl , the Ca^{2+} ions will be absorbed in the lattice and occupy the sites of Na^+ ions. If nothing else happened at the same time, the effect of this would be to upset the electric neutrality. One can assume that the charge disturbance is compensated by Cl^- ions simultaneously occupying interstitial sites. It is also possible, however, that the Cl^- ions occupy only their proper lattice sites, but that somewhere an Na^+ ion is missing. Our list of lattice imperfections can thus be extended to include:

- 5) divalent positive ion + negative interstitial ion,
- 6) divalent positive ion + positive-ion vacancy.

Which of these two imperfections occur in reality will appear in the following.

Ionic conduction and dielectric losses in NaCl doped with CaCl_2

The investigations about to be described refer to the case just mentioned in which small quantities of CaCl_2 (of the order of 0.01 atomic per cent) were added to NaCl crystals. According to the foregoing, each Ca^{2+} ion must be associated either with *a*) an interstitial Cl^- ion or *b*) an Na^+ vacancy. Conductivity measurements ⁸⁾ had already indicated that the second possibility was more favourable, and this

⁷⁾ The compounds CsCl , CsBr and CsI have a different, although also cubic, structure.

⁸⁾ See e.g. H. W. Etzel and R. J. Maurer, *J. chem. Phys.* **18**, 1003, 1950.

was confirmed by accurate examinations of the density of mixed crystals by Pick and Weber⁹⁾. They found that mixed crystals of KCl-CaCl₂ do not increase in density with increasing CaCl₂ content, as one would expect if an interstitial Cl ion accompanied each Ca ion added. They found, on the contrary, that the density decreased with increasing CaCl₂ content, pointing to the creation of vacancies.

What now is the situation with regard to ionic conduction? We have already seen that vacancies can change their position owing to an ion occupying the vacant site and thus leaving its own site vacant. In occupying a vacant site a positive ion moves on the average in the direction of the field, while the vacancy moves in precisely the opposite direction. The Na vacancies therefore behave as if they were negatively charged particles.

If β is the mobility of the vacancy (ratio of the mean velocity \bar{v} to the field strength E), n the concentration of the vacancies (ratio of the number of vacancies to the volume of the crystal) and e the "charge" of the vacancy (= electronic charge), the conductivity is given by

$$\gamma = n e \beta, \quad \dots \quad (1)$$

since, for a field E , the current density is $n e \bar{v}$, which is $n e \beta E$.

If an alternating electric field be applied, the current will then no longer lead the field by 90°, as does the pure displacement current in an ideal insulator, but instead leads the field by an angle <90° owing to the conduction which gives a component in phase with the field. As a result a certain amount of energy is dissipated in the medium in every current cycle, in other words dielectric losses occur.

For an alternating field $E = E_m \exp(j\omega t)$ we can at once write down the mean displacement \bar{x} of a vacancy, for

$$\bar{v} = \frac{d\bar{x}}{dt} = \beta E = \beta E_m \exp(j\omega t). \quad \dots \quad (2)$$

Hence

$$\bar{x} = \frac{B}{j\omega} E_m \exp(j\omega t) = -j \frac{\beta}{\omega} E. \quad \dots \quad (3)$$

The dielectric displacement $D = \epsilon E$, where ϵ is the dielectric constant, is given by

$$D = \epsilon_1 E + n e \bar{x} = \epsilon_1 E - j \frac{\gamma}{\omega} E. \quad \dots \quad (4)$$

The term $\epsilon_1 E$ is the contribution from the

"medium" *per se*, i.e. the dielectric displacement in the ideal crystal (without vacancies). The dielectric constant ϵ is now:

$$\epsilon = \frac{D}{E} = \epsilon_1 - j \frac{\gamma}{\omega}, \quad \dots \quad (5)$$

in which the imaginary term, γ/ω , determines the dielectric losses.

May we now assume that n is equal to the concentration of the added Ca ions? That would mean that all vacancies were able to move freely. We have seen, however, that an Na vacancy behaves like a negative particle. Owing to its double charge, the Ca ion exhibits a local excess of positive charge. The vacancy will therefore be attracted by the Ca ion and will tend to occupy the site of one of the twelve Na ions surrounding the Ca ion. The vacancy is then still at liberty to occupy any arbitrary one of the twelve sites, but it will not readily be able to break away from the Ca ion, at least not at low temperatures. This situation is illustrated in *fig. 1*, in which *A* represents a Ca ion, *B* a vacancy bound to this ion and *C* a free vacancy. It is reasonable to assume that at room temperature, considerable num-

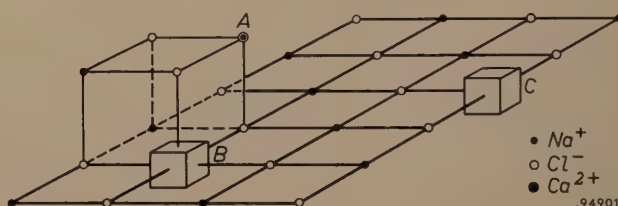


Fig. 1. Part of an NaCl lattice with an incorporated Ca²⁺ ion (*A*). *B* represents an Na vacancy bound to Ca²⁺ and *C* a free vacancy.

bers of vacancies are bound to Ca ions and therefore can no longer participate in the conduction. The chief problem now is to determine the ratio between the number of free and the number of bound vacancies. Closer examination of this problem leads to interesting theoretical conclusions, which we shall deal with here in some detail.

The Ca ion with a vacancy bound to it behaves like a "dipole" that can be aligned in a constant electric field. This alignment does not take place immediately after a field is applied but only after a certain delay. We may assume, as Debye did in the case of dipoles in liquids, that the total electrical dipole moment changes according to a time function

$$1 - \exp(-t/\tau).$$

The symbol τ represents a relaxation time, which is a measure of the "friction" to which the dipole is subjected in the "medium". The greater this friction, the greater is τ . Regarded atomically, τ is

⁹⁾ H. Pick and H. Weber, Z. Phys. 128, 409, 1950.

proportional to the average time that an Na ion must wait before it has an opportunity to move to an adjacent bound vacancy, in the absence of an external field¹⁰). When an alternating field is applied, the dipoles will change their orientation periodically under the influence of this field. In the first place this affects the dielectric constant of the crystal. The orientation of the dipoles will not, however, be in phase with the alternating field, but will lag in phase. Thus there will again be an imaginary contribution to the dielectric constant, giving rise to dielectric losses.

For the dielectric constant ϵ of a medium showing dielectric losses we can write:

$$\epsilon = \epsilon' - j\epsilon'', \dots\dots\dots (7)$$

in which ϵ' and ϵ'' are both positive. According to the work of Debye

$$\epsilon' = \epsilon_1 + \frac{a}{1 + \omega^2\tau^2}, \dots\dots\dots (8)$$

$$\epsilon'' = \frac{\gamma(0)}{\omega} + \frac{a\omega\tau}{1 + \omega^2\tau^2}, \dots\dots\dots (9)$$

$$\gamma = \epsilon''\omega = \gamma(0) + \frac{a\omega^2\tau}{1 + \omega^2\tau^2}, \dots\dots\dots (10)$$

where γ is now the A.C. conductivity, and

$$\gamma(0) = n_v e \beta \dots\dots\dots (10a)$$

is the D.C. conductivity, earlier denoted γ (conductivity at zero frequency), which is attributable to free vacancies, having a concentration n_v .

The constant a is proportional to the concentration n_d of the dipoles (bound vacancies). According to Debye's theory,

$$a = \frac{n_d p^2}{3kT}, \dots\dots\dots (11)$$

where k is Boltzmann's constant and p the dipole moment. If d is the Na-Na distance in the lattice and hence the distance between a Ca ion and a neighbouring Na vacancy, we have

$$p = e d. \dots\dots\dots (12)$$

Equations (8), (9) and (10) can each in themselves serve to define the dielectric behaviour of the crystal. We can also introduce the loss angle δ , whose tangent

$$\tan \delta = \frac{\epsilon''}{\epsilon'} \approx \frac{\gamma(0)}{\epsilon_1 \omega} + \frac{a}{\epsilon_1} \frac{\omega \tau}{1 + \omega^2 \tau^2}. \dots\dots\dots (13)$$

¹⁰⁾ For the calculation of τ see: Y. Haven and J. H. van Santen, Dielectric relaxation of lattice defects in crystals, Suppl. Nuovo Cimento 7, 605-611, 1958 (No. 2).

The ω -dependent term of ϵ' (2nd term in (8)) is here neglected, which is permissible because in the present case $a/\epsilon_1 \ll 1$. Since ϵ_1 is a constant, we have that $\tan \delta$ is, to a first approximation, proportional to ϵ'' .

In fig. 2 the theoretical variation of $\tan \delta$ as a function of $f = \omega/2\pi$ is plotted in a double logarith-

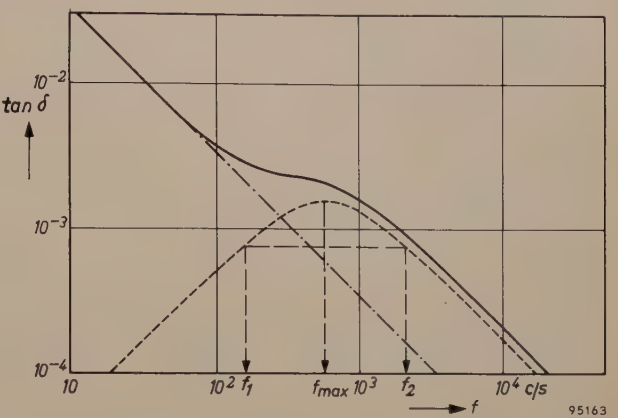


Fig. 2. Theoretical curve of $\tan \delta$ for a crystal exhibiting conduction by free vacancies and dipoles. The dot-dash line represents the contribution of the free vacancies, and the dashed line that of the dipoles. The latter curve is symmetrical with respect to f_{\max} , the frequency of the maximum. At f_1 and f_2 the ordinate is half of that of the maximum; $\log f_2/f_1$ is then a measure of the width of the maximum which is theoretically 1.14.

mic graph. The contributions of the two terms of (13) are also plotted separately. The first term, which is due to the free vacancies, is a straight line with a slope of 45°; the second term, which represents the effect of the dipoles is a symmetrical curve. The maximum of this latter term occurs when $\omega\tau = 1$, so that $\omega_{\max} = 2\pi f_{\max} = 1/\tau$.

We shall now inquire into how far the behaviour of the mixed crystals of NaCl-CaCl₂ is consonant with the theory outlined above. For this purpose, we refer to fig. 3 in which the measured value of $\tan \delta$ is plotted in a double logarithmic graph as a function of f , the frequency¹¹).

We see that $\tan \delta$ does in fact exhibit the expected behaviour as soon as a small quantity of CaCl₂ is added to NaCl, whereas in the case of pure NaCl (bottom curve in fig. 3) no such behaviour is observed. It can also be seen that the losses increase with increasing CaCl₂ content.

If we compare the curves for 0.03 and 0.14 at. % CaCl₂, we see that the inflexion in the curves shifts upwards but *not* sideways as the concentration increases. This lends weight to the assumption

¹¹⁾ Y. Haven, Dielectric losses of sodium chloride crystals, J. chem. Phys. 21, 171-172, 1953. See also Y. Haven, Dielektrische verliesen van kristallen, Chem. Weekbl. 51, 15-20, 1955.

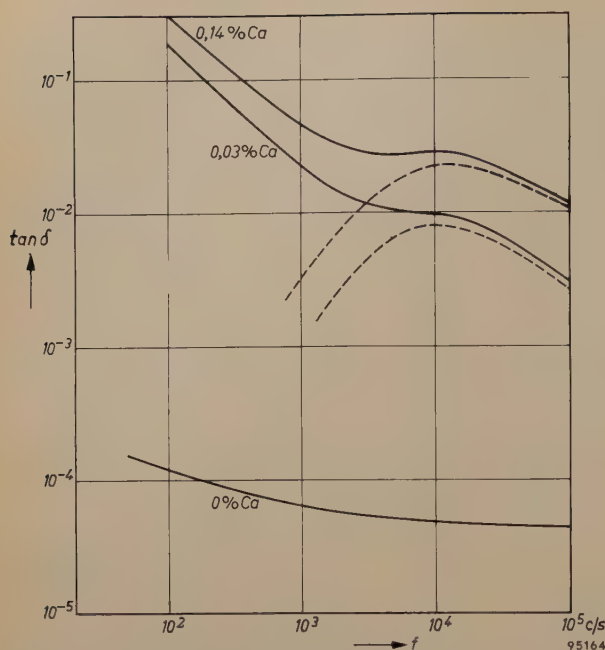


Fig. 3. Measured curves of $\tan \delta$ in NaCl and in mixed crystals of NaCl-CaCl₂. The ratio Ca/Na is given as atoms per cent.

that the inflexion in the curve is due to dipoles. This assumption is also borne out by the fact that the width of the curve representing the dipole contribution (see caption, fig. 2) is more or less in agreement with the width theoretically expected. The width found was 1.2 to 1.3 times the value expected. This is presumably attributable to subsidiary effects neglected in the simplified theory.

Fig. 4 shows the behaviour of the losses as a function of temperature for a specific CaCl₂ content. The maximum of the broken curve (dipole contributions) shifts to the right, i.e. to higher frequencies with increasing temperature. Since $\omega_{\max} = 1/\tau$, this means that τ decreases with rising temperature, that is, that the rotational mobility of the dipole increases, as was to be expected. The height of the maximum (determined by the value of a , and hence that of n_d) also rises slightly. We shall return to this

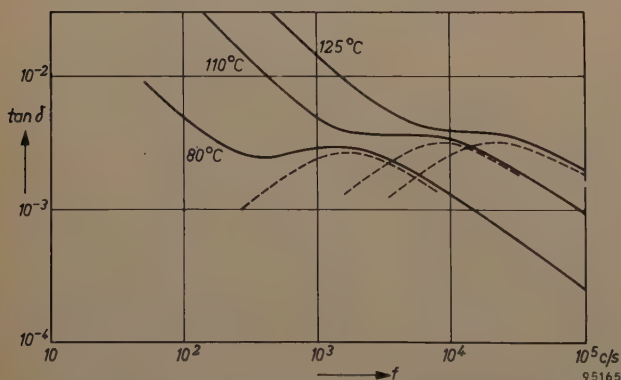
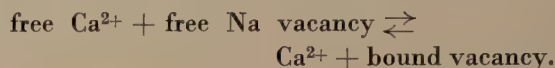


Fig. 4. Curves of $\tan \delta$ against frequency f in a mixed crystal of NaCl-CaCl₂ at different temperatures. The broken lines represent the dipole contributions.

effect later. On the basis of the picture outlined above the sum of the concentration n_v of the free vacancies and the concentration n_d of the dipoles is equal to the concentration n_{Ca} of the Ca ions added to the crystal:

$$n_v + n_d = n_{Ca}.$$

It can be shown that we can reasonably assume this to be the case at moderately high temperatures ($\sim 1000^\circ\text{K}$). In that case n_v and n_d can, in principle, be determined from the chemical equilibrium:



In this reaction we can apply the law of mass action:

$$\frac{n_{\text{free Ca}} \times n_v}{n_d} \approx \frac{n_v^2}{n_d} = K, \quad \dots \quad (14)$$

where K is the equilibrium constant (dependent on the temperature) for the reaction, given by

$$\ln K = C - \frac{q}{kT}, \quad \dots \quad (15)$$

in which q is the "dissociation energy" per dipole and C is again a constant, related to the Ca concentration. Hence with increasing temperature, K increases also. For $T = 1000^\circ\text{K}$ we estimate $n_v/(n_v + n_d) = n_v/n_{Ca} \approx 0.8$, showing that at this temperature most of the vacancies are free vacancies.

Putting $n_{Ca}/K = z$ and introducing the "degree of dissociation" $a = n_v/n_{Ca}$, we may write equation (14) in the form

$$\frac{a^2 z}{1-a} = 1. \quad \dots \quad (14a)$$

This leads to

$$\frac{n_v}{K} = az = -\frac{1}{2} + \sqrt{z + \frac{1}{4}}, \quad \dots \quad (16)$$

and

$$\frac{n_d}{K} = (1-a)z = z + \frac{1}{2} - \sqrt{z + \frac{1}{4}}. \quad \dots \quad (17)$$

In the case of (16), we see that $az \approx z$ when $z \ll \frac{1}{4}$, and $az \approx \sqrt{z}$ when $z \gg \frac{1}{4}$. For (17) we have $(1-a)z \approx z^2$ when $z \ll \frac{1}{4}$ and $(1-a)z \approx z$ when $z \gg \frac{1}{4}$. Both functions are shown in fig. 5. This figure also shows the curve of a as a function of z . For $z \ll \frac{1}{4}$, $a \approx 1-z$, and for $z \gg \frac{1}{4}$, $a \approx 1/\sqrt{z}$. As $z \rightarrow \infty$ the value of a slowly approaches zero. From this we see that at high temperature (large K) and not too large n_{Ca} , the value of a approaches unity and hence $n_v \approx n_{Ca}$. The dissociation is then practically complete. At low temperature (small K) and large n_{Ca} we find $1-a \approx 1$ or $n_v/n_{Ca} \approx 0$, and hence $n_d \approx n_{Ca}$, which means that there is almost complete association.

Having estimated in this way the value of n_v (strict accuracy being neither necessary nor possible), we can now derive from the D.C. conductivity $\gamma(0) = n_v e \beta$ the mobility β of the free vacancies. Clearly, β , like $\gamma(0)$, is very dependent on temperature.

At the low temperatures, e.g. about 100 °C, at which our experiments were performed, one is no longer so certain whether the above-mentioned condition $n_v + n_d = n_{Ca}$ is satisfied or whether the equilibrium discussed gives a true picture of the

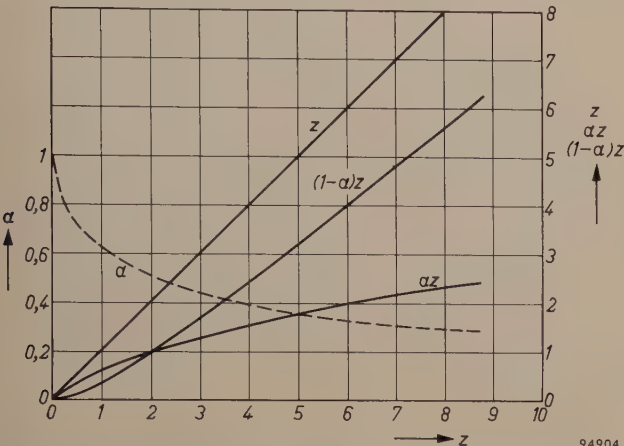


Fig. 5. Graph of αz , $(1 - \alpha)z$ and α as a function of $z = n_{Ca}/K$, to illustrate the chemical dissociation equilibrium $\alpha^2 n_{Ca}/(1 - \alpha) = K$ (see eq. 14a).

situation in the crystal. One can then, however, acquire information about n_v and n_d independently.

On the assumption that equation (11) is correct, we can determine n_d from the value of α , i.e. from the $\tan \delta$ curve.

As regards n_v , we earlier calculated its value (approximately) at high temperatures from equations (14) and (15) and derived the values of β from the measured values of $\gamma(0)$ at those temperatures; now at low temperatures we can conversely derive n_v from the value of $\gamma(0)$ measured at these low temperatures. This value is, of course, found by extrapolating $\omega \tan \delta$ to $\omega = 0$. To find n_v , however, we must also know the value of β at low temperatures. Now, at high temperatures, the relation between β and T can be represented by an empirical formula of the form:

$$\beta = \frac{A}{T} \exp \left(-\frac{B}{T} \right) \quad (18)$$

The value of β at low temperatures can thus be estimated by extrapolating the curve corresponding to (18) to the temperatures in question.

No more than a rough estimate is obtained in this way: the values of A and B reported by various investigators show quite considerable disparities, indicating that extrapolation to low temperatures involves a rather high degree of uncertainty. In Table I are given the values of A and B as found by different investigators, and also the corresponding values of β (in units of $(\text{m/s})/(\text{V/m}) = \text{m}^2/\text{Vs}$) for $T = 1000^\circ\text{K}$ and 383°K .

Table I. Values of A and B in (18) as estimated by various investigators, and the associated values of β at 1000 °K and 383 °K¹². The symbol ν represents the average frequency (derived from β) at which Na^+ ions jump to free vacancies (at 383 °K).

Investigator	A	B	$\beta \text{ (m}^2/\text{Vs)}$		ν
			1000 °K	383 °K	
Etzel-Maurer	1.96	9860	10.5×10^{-8}	3.4×10^{-14}	3.5×10^3
Lidiard	2.12	9750	12.8	5.2	5
Bean	0.46	9050	5.5	7.1	7
Haven	0.225	8360	5.2	20	21

It can be seen that at 1000 °K the values of β differ by a factor of about 2, and at 383 °K by a factor of about 6.

In Table II second column, are set out the number of free Na vacancies per Na^+ ion (n_v/n_{Na}) calculated from the author's measurements of $\gamma(0)$ and β , as a function of the total number of Ca^{2+} ions per Na^+ ion (n_{Ca}/n_{Na}) given in the first column. The fourth column gives the number of dipoles (n_d/n_{Na}) calculated from the dielectric losses.

Table II. Number of free and bound vacancies per Na^+ ion (2nd and 4th columns) in mixed crystals of NaCl-CaCl_2 , as a function of the total number of Ca^{2+} ions per Na^+ ion. The figures in the third and fifth columns indicate the vacancies as a fraction of Ca^{2+} .

n_{Ca}/n_{Na}	Free Na vacancies (n_v)		Bound Na vac. (n_d)	
	n_v/n_{Na}	n_v/n_{Ca}	n_d/n_{Na}	n_d/n_{Ca}
0.6×10^{-4}	0.003×10^{-4}	0.005	0.08×10^{-4}	0.13
1.0	0.016	0.016	0.16	0.16
1.7	0.028	0.016	0.60	0.35
2.5	0.043	0.017	0.9	0.36
3.6	0.07	0.019	1.5	0.42
5.2	0.11	0.021	3.2	0.62
9	0.13	0.014	3.0	0.33
17	0.19	0.011	6.2	0.36
27	0.3	0.011	6.7	0.25

In fig. 6 the values of n_v/n_{Na} and n_d/n_{Na} from Table II are plotted against n_{Ca}/n_{Na} . The values of n_v/n_{Na} have been multiplied by 10, partly for reasons of clarity in the graph and partly to allow for a possibility that the values of β used in our calculation might have been too large (see author's value Table I). The sum $n_d/n_{Na} + 10 n_v/n_{Na}$ is plotted as the broken curve. It may reasonably be assumed that this curve represents an upper limit to the values of $(n_v + n_d)/n_{Na}$.

¹² Y. Haven, Concentration and association of lattice defects in NaCl, Rep. Conf. on defects in crystalline solids (Bristol 1954), 1955, pp. 261-272.

If the situation is completely in accord with the equilibrium ($n_v + n_d = n_{Ca}$) assumed above, we should find for $(n_v + n_d)/n_{Na}$ a 45° line. We see that, for $n_{Ca}/n_{Na} \leq 5 \times 10^{-4}$, the values found remain 20 to 40% below that line; at higher concentrations the deviation is still greater, for the values are roughly constant.

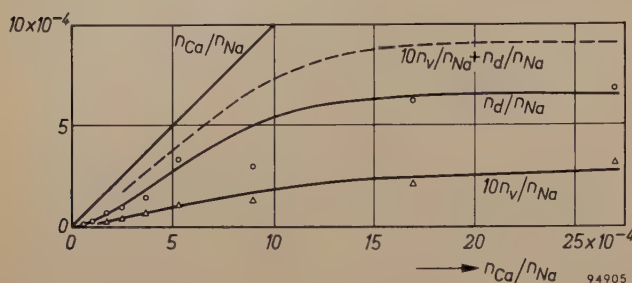


Fig. 6. Values, taken from Table II, of n_d/n_{Na} and n_v/n_{Na} (the latter $\times 10$) plotted against n_{Ca}/n_{Na} . These curves should be compared with the theoretical curves in fig. 5. The substantially horizontal parts of the curves at high Ca concentrations indicates segregation of $CaCl_2$.

The substantially constant values found at Ca^{2+} concentrations greater than about 10^{-3} is probably because, at these concentrations, the solubility limit of $CaCl_2$ in solid $NaCl$ is exceeded, in which case the surplus $CaCl_2$ is precipitated. In analogous cases, e.g. in $NaCl$ to which $PbCl_2$ or $CdCl_2$ had been added, it has been possible to observe this precipitation in that the formerly transparent crystal becomes turbid (a milky clouded appearance due to the different refractive indices of $PbCl_2$ and $CdCl_2$).

Confining our attention to the smaller concentrations ($n_{Ca}/n_{Na} < 5 \times 10^{-4}$), it would be very agreeable if we were able to compare the form of the n_v and n_d curves with the theoretical curves of fig. 5. This is not readily possible, however, partly because of the uncertain value of n_v and partly because the value of the reaction constant K at low temperature is not accurately known.

The fact that the values found for n_v and n_d are smaller than corresponds to $n_v + n_d = n_{Ca}$ does not in itself prove that the equilibrium that we assume with some justification at high temperatures is no longer valid at low temperatures. It is quite conceivable that the real values of n_v and n_d are larger than the values derived from observations. As regards n_d it may well be that a smaller value for the dipole moment p should be inserted in (11) than the value ed from (12): it is possible that the Ca ions in their holes are slightly attracted towards the Na vacancies, so that the length of the dipole becomes smaller than d . If we do not assume that the too low values derived for n_v and n_d are entirely attributable to such an effect, we must necessarily

conclude that yet other combinations of Ca^{2+} ions and vacancies are possible at low temperatures, i.e. more complex configurations which do not contribute to the dielectric losses. The increase in $(\tan \delta)_{\max}$ with increasing temperature, to which reference has already been made (fig. 4), might point to this: at higher temperatures the more complex configurations become dissociated.

In this connection it should be pointed out that the above conjecture that the Ca^{2+} -vacancy spacing is less than the normal Na^+ - Na^+ spacing d might conceivably go so far that the spacing becomes $\frac{1}{2}d$. In that case the Ca^{2+} ion is interstitially positioned between two Cl^- ions, as illustrated in fig. 7. One can also regard this as a $CaCl_2$ molecule that has replaced two $NaCl$ molecules, with the result that two vacancies are created, one on each side of the Ca ion. Such a configuration would have zero dipole moment.

It is also conceivable that more complex configurations exist which do have a dipole moment, and hence also contribute to the losses, but for which the relaxation time τ differs from that applicable to the simple dipoles, so that ω_{\max} (fig. 2) has a different value. In fact some loss-curves measured at low temperatures give indications of extra maxima outside the principal maximum caused by the dipoles.

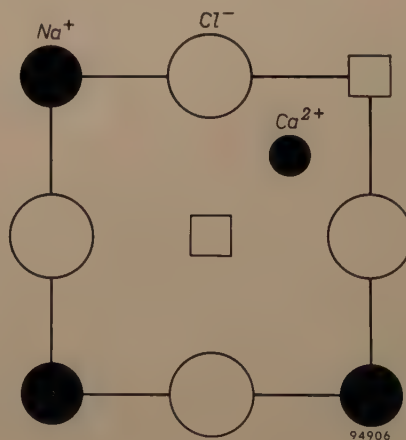


Fig. 7. Possible interstitial position of a Ca^{2+} ion between two Cl^- ions. The squares represent Na vacancies. This configuration has zero dipole moment.

Finally it should be mentioned that in silver halides, e.g. in mixed crystals of $AgBr$ - $CdBr_2$, dipole concentrations n_d have been found that are substantially lower than the concentration of foreign ions, namely about 60% lower (on the assumption that the dipole moment is given by ed ¹³).

¹³) J. Teltow and G. Wilke, Naturwiss. 41, 423, 1954.

The mechanism of the movement of vacancies

As we have seen above, ionic conduction takes place both in pure NaCl and in mixed crystals of NaCl and CaCl_2 owing to Na ions moving to neighbouring vacancies and in their turn leaving a vacant site behind. With AC conduction in NaCl- CaCl_2 the vacancies that are bound to Ca ions also contribute to the conduction; the Na ion that fills such vacancies then becomes one of the positive neighbours of the Ca ion.

The question now arises in what way an Na ion, whether "free" or "bound" moves through the lattice to an Na vacancy. At all events the Na ion has to force its way between the surrounding Cl ions, that is to say it must cross a potential barrier. To do so it requires a certain activation energy which it can draw from the thermal agitation.

An Na ion is surrounded in the crystal lattice by six Cl ions which form the corners of an octahedron, as illustrated in perspective by *fig. 8*. The Cl ions *A* and *B* lie in a plane parallel to the plane of the drawing (a (110) lattice-plane, diagonal plane of the cubic unit cell), and the ions *C*, *D*, *E* and *F* lie in front and behind this plane at distance of $\frac{1}{2}\text{Cl-Cl}$. Two extra Cl ions are shown, namely *G* and *H*, also in the first-named plane.

At *K* there is an Na ion and at *L* a vacancy. The Na ion can move to the vacancy in a straight line via point *1* (midway between *C* and *D* in the 110 plane), or, as shown by the broken line, via points 2

(midway between three Cl ions) and 3 (midway between four tetrahedral Cl ions), or again it can follow a path somewhere between these extremes, for example via point 4, which lies just as far from the Na ion *M* as does point *K*. In considering which of these paths is most probable, we must take into account mainly the following contributions to the potential energy:

- 1) Coulomb energy,
- 2) polarization energy, and
- 3) repulsion energy.

It has not yet proved possible to calculate the absolute height of the potential barrier for the various paths, but the above-mentioned measurements of dielectric losses and the electrical conductivity permit some qualitative statements to be made.

We are interested in the first place in the difference between the jump to a free vacancy and the jump to a bound vacancy. The most important factor here is the repulsion which the jumping Na ion suffers from the extra positive charge of Ca^{2+} , i.e. the difference in the Coulomb energy contribution in consequence of the presence of Ca^{2+} .

At *M* in the figure an Na ion is shown that also lies at a distance of $\frac{1}{2}\text{Cl-Cl}$ or $\frac{1}{2}\text{Na-Na}$ in front of the plane of the drawing. We now imagine this ion to be substituted by Ca^{2+} . This gives a Coulomb contribution to the potential energy amounting, for the points indicated in the figure, to:

<i>K</i> or <i>L</i>	<i>1</i>	<i>2</i>	<i>3</i>	<i>4</i>
3.592	4.165	3.326	3.064	3.592 eV

Movement of the Na ion in from *K* (or *L*) to the positions 1, 2, 3 or 4 thus involves the following changes (due to Coulomb interaction *alone*) in the required activation energies:

<i>1</i>	<i>2</i>	<i>3</i>	<i>4</i>
+0.57	−0.27	−0.53	0 eV

The actual net change in potential energy is probably smaller, since the polarization energy has a compensating effect, but we may safely assume that, owing to the presence of the Ca, the already difficult path via *1* becomes even more difficult, and that via *2* and *3* easier.

This is more or less in agreement with what we may conclude from the observations about the jump frequencies. The jump frequency of Na^+ to a bound vacancy at 110 °C can be calculated as 40×10^3 per sec. On the other hand the value of the jump frequency ν to a *free* vacancy can be derived from

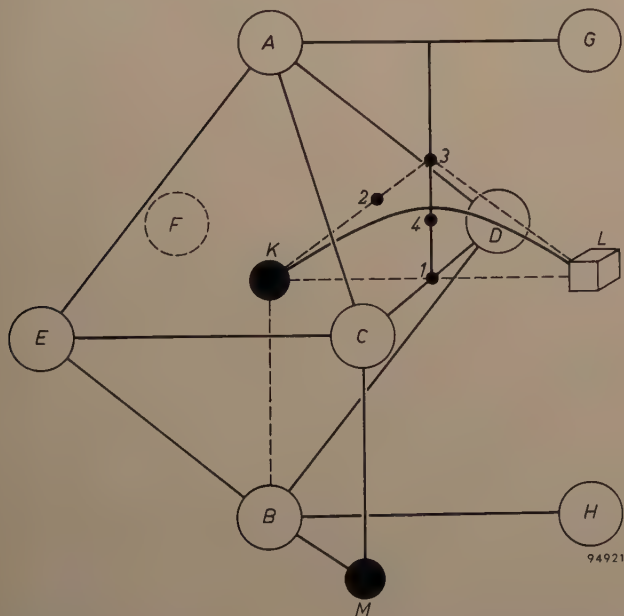


Fig. 8. Perspective sketch of a part of an NaCl crystal, with a (110) plane (diagonal plane of cube) parallel to the plane of the drawing. *A*...*H* represent Cl ions, *K* an Na ion, *L* a vacancy, *M* an Na ion that can be replaced by a Ca ion. The possible paths of an Na ion are indicated (via *1*, via *2* and *3*, and between *3* and *4*).

the quantity β ¹⁴). The values found, varying from 3.5×10^3 to 21×10^3 per sec, are given in Table I.

Comparison of the above values for the jump frequencies, incomplete as they may be, indicate that the potential barrier is somewhat lowered in the presence of a Ca^{2+} ion. It is reasonable to assume that the most favourable path for the Na^+ ion is a bent path passing, for example, between points 3 and 4 in the figure. This applies both for the jump to a free and to a bound vacancy, for we cannot expect the route to be greatly altered by the presence of Ca^{2+} .

Ionic conduction in silver-halide crystals

In connection with the results discussed above, we shall now briefly mention some particulars concerning silver-halides. Since these are also monovalent compounds one might be inclined at first sight to believe that conduction takes place in these crystals in the same way as in the alkaline halides, namely by means of free vacancies (Schottky defects). In fact, results resembling those for alkaline halides are obtained when a Cd halide is added as an impurity to Ag halides. An Ag^+ ion is then replaced by Cd^{2+} giving rise to Ag^+ vacancies, which cause conduction. In the pure halides AgCl and AgBr, however, there is evidently a preference for a different kind of lattice imperfection, viz. Frenkel defects, whereby Ag vacancies are associated with interstitial Ag ions (case 1 of the section dealing with lattice imperfections in alkaline halides). In this case, too, of course, the vacancies can cause conduction. However, the interstitial atoms also contribute to the conduction, and since these are approximately four times as mobile as the vacancies, conduction in the pure compounds is mainly due to the interstitial atoms.

Conduction due to interstitial atoms is an indirect effect. One must assume that an interstitial atom displaces a regular lattice atom and occupies its site, and that the displaced atom is driven to an interstitial site, from which the process can be repeated.

The latter conclusion was drawn by comparing the diffusion constant of Ag ions in the crystal with its contribution to the ionic conductivity.

Colour centres in mixed crystals of NaCl-CaCl₂

Finally, returning to the mixed crystals of NaCl-CaCl₂ which we have already considered in some detail, we shall now consider what happens in these

crystals when, besides the existing vacancies, other lattice imperfections, namely colour centres, are present.

When a pure alkali-halide crystal is heated in the vapour of its alkali metal, e.g. an NaCl crystal in Na vapour or a KCl crystal in K vapour, the originally clear crystal is seen to become coloured. KCl, for instance, turns purple and exhibits in its absorption spectrum an absorption band in the green.

In accordance with a conjecture of J. H. de Boer dating back to 1935, it is assumed that a K atom attaches itself as an ion to the crystal and unites with a Cl ion diffusing out of the lattice. The electron of the K atom moves to the vacancy left by the departure of the Cl ion. This electron, which gives rise to the absorption band, is called a colour centre (*F* centre)¹⁵. It has been uncertain, however, whether the electron occupies the middle of a vacancy (Mott) or whether it unites more or less with the positive ions surrounding the vacancy (De Boer). The greater likelihood of the latter view has been confirmed, among other things, by investigations of the paramagnetic resonance of *F* centres in alkali-halide crystals¹⁶.

Colour centres can also be brought about in the crystal at high temperatures when electrolytic conduction takes place under a strong electric field. The electrons that must enter the lattice in order to form the colour centres originate in this case directly from the negative electrode.

When the above-discussed mixed crystals of NaCl and CaCl₂ are heated in Na vapour, not only are colour centres formed, but the divalent Ca ions (Ca^{2+}) may be reduced to monovalent ions (Ca^+). Further to what has been said above on pure alkali halides, one can regard this as a process in which electrons are introduced into the crystal, some of which do not move to a Cl vacancy but attach themselves to a Ca^{2+} ion. It should be noted, however, that most of the electrons find a place in the Cl vacancies and so give rise to the usual colour centres. Large numbers of monovalent Ca^+ ions are only obtained when the crystal, treated as described, is irradiated at lower temperatures by light in the absorption band of the colour centres. This might be explained on the following basis. Irradiation in the *F* band raises the electron in the vacancy to a higher energy level; as a result the electron is able to break its bond with the vacancy, and thereafter

¹⁴) The jump frequencies of the bound and free vacancies themselves are respectively 4 and 12 times higher, in view of the number of Na^+ neighbours present in each case.

¹⁵) See e.g. E. J. W. Verwey, Electronic conductivity of non-metallic materials, Philips tech. Rev. 9, 46-53, 1947/48.

¹⁶) See e.g. J. S. van Wieringen, Paramagnetic resonance, Philips tech. Rev. 19, 301-313, 1957/58 (No. 11).

to move more or less freely through the lattice. If in so doing it meets a Ca^{2+} ion, it enters into union with it.

This phenomenon can be demonstrated very elegantly in the analogous case of KCl to which SrCl_2 has been added. Here, too, F centres are chiefly formed (by heating in K vapour) as a result of which the crystal, as before, turns purple. After cooling, the crystal is irradiated with green light, whereby the colour changes to red and a new absorption band in blue is formed, pointing to a somewhat stronger bond. This absorption is attributable to the formation of Sr^+ ions, which can be regarded as a complex of $\text{Sr}^{2+} + \text{electron}$ situated in the lattice.

The phenomenon of divalent ions being reduced to monovalent ions has some connection with the phenomenon of "controlled" valency¹⁷⁾. In the case described above we were concerned with divalent ions incorporated in a lattice of monovalent ions. Now let us see what happens when, conversely, monovalent ions are incorporated in a lattice of divalent ions, e.g. Li_2O added to NiO . One might imagine that here, too, electric neutrality would be preserved by the occurrence of vacancies, viz. O vacancies. It is found in this case, however, to be energetically favourable if, for each incorporated Li^+ ion, an Ni^{2+} ion is simultaneously oxidized to Ni^{3+} . The lattice imperfections occurring here thus consist in the substitution of two Ni^{2+} ions by a monovalent and a trivalent ion.

The causes of the differing behaviour of the two kinds of mixed crystals, NaCl-CaCl_2 and $\text{NiO-Li}_2\text{O}$ can be described in the following way. The electron that reduces the Ca^{2+} ion to Ca^+ is admittedly bound somewhat more strongly to the Ca^{2+} ion than in an F centre, but the very large number of possible ways in which the second case, the combination $\text{Ca}^{2+} + \text{Na vacancy} + F$ centre, may be realized compared with the first case, $\text{Ca}^{2+} + e \rightarrow \text{Ca}^+$, swings the balance in favour of the second case ("entropy gain" prevails over "energy gain"). The energy gain in the bound with Ca^{2+} is particularly small in this case owing to the loss of polarization energy during the formation of the bond.

In the $\text{NiO-Li}_2\text{O}$ system, on the other hand, it is advantageous for an electron to be withdrawn from Ni^{2+} , because the polarization energy here contributes positively to the process, and moreover a comparison of the probability of occurrence of $2\text{Li}^+ + 2\text{Ni}^{3+}$ with that of $2\text{Li}^+ + \text{O vacancy}$ shows that the entropy gain in the former process is larger.

Concluding remarks

The investigations described in the foregoing have led to a quantitative insight into a number of phenomena which must be attributed to the presence of lattice imperfections in crystals of alkali halides and in silver halides. The investigations have in the first place increased our knowledge of the behaviour of these relatively simple compounds, but their principal importance lies in the fact that they point the way to the explanation of the behaviour of more complex crystals, for which alkali and silver halides can serve as a "model". Compound lattice imperfections in certain crystals, interesting perhaps because of their technical importance, can make their influence felt in extremely complicated ways. Any attempt to analyse this influence must necessarily be preceded by careful verification of the role played by the various lattice imperfections in more elementary compounds such as the alkali halides.

Summary. After an introductory discussion of crystal structure and lattice imperfections in general, the author deals more in detail with the situation in alkaline halides, both in the pure state (Frenkel and Schottky defects) and after the introduction of impurities consisting of a halide of a divalent metal. Ionic conduction and dielectric losses in mixed crystals of NaCl-CaCl_2 are dealt with at some length with reference to measurements of dielectric loss angle $\tan \delta$ as a function of frequency. It is shown that Na^+ vacancies occur in these processes, some of these vacancies being free and others associating with Ca^{2+} ions to form dipoles. The degree of association of these dipoles is determined at higher temperatures, and possibly also at lower temperatures, by a simple chemical equilibrium. For the purpose of a quantitative agreement with the theory of this equilibrium, however, it is necessary to assume that the dipole strength may be smaller than the obvious value ed (where e is the electronic charge and d the Na-Na spacing in the lattice). It is also possible that at low temperatures there exist, besides the dipoles, more complex configurations which make no contribution, or at least very little, to the dielectric losses. After a discussion of the mechanism of ionic conduction and the path of the Na^+ ion in the lattice, the behaviour of silver halides is compared with that of alkali halides. Finally, some attention is given to the behaviour of NaCl-CaCl_2 when heated in Na vapour (formation of colour centres and reduction of Ca^{2+} to Ca^+).

¹⁷⁾ E. J. W. Verwey and F. A. Kröger, New views on oxidic semi-conductors and zinc-sulphide phosphors, Philips tech. Rev. **13**, 90-95, 1951/52.

ABSTRACTS OF RECENT SCIENTIFIC PUBLICATIONS BY THE STAFF OF N.V. PHILIPS' GLOEILAMPENFABRIEKEN

Reprints of these papers not marked with an asterisk * can be obtained free of charge upon application to the Philips Research Laboratory, Eindhoven, Netherlands.

2544: P. C. van der Willigen: De invloed van waterstof op de kwaliteit van de las (De metalurgie van het lassen van staal, Lassymposium 1957, Ned. Ver. voor Lastechniek, The Hague, pp. 24-29). (The effect of hydrogen on the quality of the weld; in Dutch.)

Although the embrittling of steel by hydrogen has been known for over a century, it is only since world war II that the unfavourable influence of hydrogen on the arc welding of steel is understood. Hydrogen is one of the causes of porosity in the weld, of microcracks in quenched weld-metal and of weld-metal cracking and underbead-cracking in low-alloy steel. A number of experiments which give an idea of the typical behaviour of hydrogen in steel are reviewed. Welding experiments on the influence of hydrogen are described, showing clearly that the danger of hydrogen, still doubted by some, is indeed very real. An attempt is made to contribute to an understanding of the phenomena.

2545: G. Diemer, G. J. van Gurp and H. J. G. Meyer: The nature of the edge emission in CdS (*Physica* **23**, 987-988, 1957, No. 10).

Letter presenting experimental evidence in confirmation and extension of an earlier view that CdS emission bands in the region of 5100-5500 Å are due to recombination of electrons and holes via impurity centres.

2546: N. W. H. Addink: A note on the determination of the Avogadro number (pp. 221-223 of supplement No. 1 to *Nuovo Cimento* **6**, series X, 1957).

Note on the small deviations in the values of Avogadro's number found by different experimental methods. The following are among the causes of the deviations: crystal imperfections and errors in density measurements, especially for small crystals. From measurements on KCl crystals prepared from the melt it is suggested that these crystals are left with $\sim 2 \times 10^{-4}$ frozen-in vacancies per cm³, which agrees satisfactorily with the number derived from conductivity measurements (1.2×10^{-4} per cm³).

2547: G. Meijer: The influence of light quality on the flowering response of *Salvia accidentalis* (*Acta bot. neerl.* **6**, 395-406, 1957, No. 4).

Salvia accidentalis is an obligate short-day plant. However, flower initiation may occur after a long-day treatment, depending on light intensity and the colour of the main light period. In *Salvia accidentalis* as in *Hyoscyamus niger* the effect of a long-day treatment is influenced by at least two different photoreactions.

2548: G. Meijer and R. van der Veen: Wavelength dependence of photoperiodic responses (*Acta bot. neerl.* **6**, 429-433, 1957, No. 4).

It has been shown in several plants that the effect of a short-day treatment does not depend on the light quality of the main light period. However, to obtain a long-day effect by interrupting the long-night period with a red or green irradiation, it is shown that for certain plant species a special blue-light requirement for the main light period exists.

2549: P. Massini: Photosynthetic phosphorylation as an explanation for induction phenomena in photosynthesis (*Acta bot. neerl.* **6**, 434-444, 1957, No. 4).

The initial uptake of CO₂ by photosynthetically active tissue after the transition from dark to light is investigated. It is suggested that this phenomenon is caused by a shift in the pH of the cell medium toward alkalinity which is due to the formation of energy-rich phosphates in the light.

2550: J. H. Stuy: Herstel van beschadiging door straling bij micro-organismen (*Chem. Weekblad* **53**, 621-625, 1957, No. 47). (Recovery of micro-organisms from radiation damage; in Dutch.)

A short survey is given of the radiation killing of micro-organisms and the subsequent reactivation phenomena. A possible connection between these phenomena is discussed.

- 2551:** J. B. de Boer: Blendung beim nächtlichen Strassenverkehr (Zentralblatt Verkehrs-Medizin, Verkehrs-Psych. angr. Geb. **3**, 185-203, 1957, No. 4). (Glare in road traffic at night; in German.)

The night driver is troubled by the glare of two kinds of light sources, headlights and lanterns for public lighting. Glare from these sources may be either disability glare or discomfort glare. Visibility tests have shown the former to be absent if the latter is kept within satisfactory limits. Further tests have allowed quantitative conclusions to be arrived at with regard to the permissible degree of discomfort glare due to public lighting. Limitation of glare to this degree, while maintaining an adequate luminance of the road surface necessary for sufficient visibility, requires more light. This is made justifiable, however, by the efficient means nowadays available for lighting roads. On roads thus lit, drivers should use side lights only, as dipped headlights, however well they work, cause considerable discomfort from glare. Good lighting for roads carrying dense traffic can therefore only be obtained from fixed sources. The influence of colour on discomfort glare is briefly discussed.

- 2552:** A. H. Boerdijk: Apparatus for routine refraction measurements of layers of lacquers with digital reading (Rev. sci. Instr. **28**, 840-841, 1957, No. 10).

Brief description of an instrument for the rapid measurement of the refractive index of thin layers, based on measurements of the Brewster angle. The range is $n = 1.3$ to $n = 1.8$, and the accuracy is to the third decimal place.

- 2553:** L. M. Nijland and L. J. van der Pauw: The effect of heat treatment on the bulk lifetime of excess charge carriers in silicon (J. Electron. Control **3**, 391-395, 1957, No. 4).

In a recent paper G. Bemski (1956) described the decrease of the bulk lifetime of excess charge carriers in silicon found when silicon samples were quenched from temperatures lower than 550°C to room temperature, and the complete or partial restoration of the original lifetime when the samples were annealed in the same range of temperatures. The authors have studied the effects of heat treatment of silicon in the range between room temperature and 1250°C . It was found that bulk lifetime both in p- and n-type crystals made by the floating-zone method and of n-type crystals made by the Czochralski technique might be increased

by annealing at temperatures between about 300°C and 700°C . Annealing at temperatures higher than about 700°C decreased lifetime both in p- and n-type crystals. In all cases the samples were relatively slowly cooled. The mechanism of these changes in lifetime is not well understood.

- 2554:** J. A. Kok and M. M. G. Corbey: Dipoles and electric breakdown (Appl. sci. Res. **B 6**, 449-455, 1957, No. 6).

It is shown that breakdown of insulating or dielectric material may be caused either by polarizable particles or permanent dipoles. Induced dipoles as well as permanent dipoles may gather at a place of maximum stress and form a bridge. Bridges consisting of permanent dipoles may cause the gap between the valence energy band and the conduction energy band to be filled by impurity or imperfection levels. The low ionization potentials of electrons attached to permanent dipoles may facilitate breakdown. An attempt is made to reconcile Crowe's results with the authors'.

- 2555:** F. L. H. M. Stumpers: Information theory and international radio organizations (IRE Trans. on Information Theory **IT-3**, 85, 1957, No. 2).

Guest editorial (to the above Transactions) on the applications of information theory in radio communication.

- 2556:** J. A. Kok: Doorslagverschijnselen in vloeibare isolatoren (Ingenieur **69**, E179-E181, 13 Dec. 1957). (Electrical breakdown of liquid insulators; in Dutch.)

The electrical breakdown strength of insulating oil depends on the size of foreign particles, which may form bridges in a place of maximum electric stress (see No. **2507** of these Abstracts). A theoretical relation has been derived and verified with colloid suspensions of particles of known radius. Mineral oils may deteriorate if the particles unite by the process of flocculation, the occurrence of which depends on the relative magnitudes of the attractive London-Van der Waals forces acting between the particles and the repulsive forces between their ion atmospheres. The possibility of using a mineral oil as an insulator depends on the existence of an upper limit of the size of particle complexes due to the rapid fall-off of the L.-v. d. W. forces at diameters exceeding 500 \AA .

2557: J. Meltzer: Insektizide und akarizide Wirksamkeit von 2,4,5,4'-Tetrachlor-Diphenyl-Sulphon (Tedion) (Z. ang. Entomologie **41**, 58-63, 1957, No. 1). (Insecticidal and acaricidal activity of 2,4,5,4'-tetrachlorodiphenyl sulphoxide (Tedion); in German.)

Tedion does not have contact poisonous activity on insects. *Musca domestica* L., *Sitophilus granarius* L. and *Ephestia kuehniella* Zell. are not killed by contact in treated Petri dishes. *Aphis fabae* Scop. and larvae of *Leptinotarsa decemlineata* Say. and *Diataraxia oleracea* L. were not killed by contact with, or feeding on plants dipped into Tedion emulsions. *Aphis mellifica* L. is not affected by Tedion as a contact poison, applied in as high a dosage as 16 g/m². *Drosophila melanogaster* has been bred in media containing 10-100 ppm of Tedion without influence on development and number of hatched flies. Two successive generations of *Ephestia kuehniella* Zell. have been bred on rolled oats, treated with 1-100 ppm of Tedion. No influence on time of development nor on percentage of hatching was noted. A feeding experiment with addition of 500-62 500 ppm of Tedion to honey-sugar paste shows that Tedion does not affect the bee. Eggs of *Tetranychus urticae* Koch could be killed by emulsions of the extracts made from the digestive tract of the treated bees, but were not affected by an extract of the control bees. Tedion appears to be a highly specific acaricide which is safe for man and animal, and which does not affect predator and parasite insects and bees.

2558: J. Meltzer: Penetratie en werkingsduur van enkele spintoviciden (Meded. Landbouwhogeschool. Opzoekingsstat. Gent **22**, 457-464, 1957, No. 3). (Penetration and residual action of some red-spider ovicides; in Dutch.)

Of the four red-spider ovicides chlorobenside, Tedion, p-chlorophenyl-p-chlorobenzene sulphonate and p-chlorophenyl benzene sulphonate, the last one is the fastest to penetrate through the treated leaves, but also the fastest to loose activity. Only Tedion remains highly active two weeks after the treatment. The oral uptake of Tedion by the females and the larvae plays an important role in its action.

2559*: M. van Tol: Automatische regeling van kern-reactoren (Chapter, pp. 87-101, in the book: Ontwikkelingen in de moderne regeltechniek, Verslag T.W.O. Kon. Inst. Ingenieurs, Utrecht 1957). (Automatic control of nuclear reactors; in Dutch.)

This paper deals with the automatic control of small nuclear reactors where temperature variations are unimportant. As an example, the automatic control of the reactor shown at the exhibition "Het Atoom" in Amsterdam, 1957, is discussed in some detail. (See also Philips tech. Rev. **19**, 245-257 and 273-285, 1957/58, Nos. 9 and 10.)

R 332: W. Ch. van Geel, C. A. Pistorius, and B. C. Bouma: Luminescence of the oxide layer on aluminium during and after its formation by electrolytic oxidation (Philips Res. Repts. **12**, 465-490, 1957, No. 6).

Report on measurements of the luminescence occurring with an anodically deposited oxide layer on aluminium (a brief description was given in J. Phys. Radium **17**, 714-717, 1956; No. **2426** of these Abstracts). During the formation of the oxide layer a pronounced luminescence appears. When the field strength in the layer is kept constant, the luminescence L is found to increase with the thickness d according to $L = a(e^{bd} - 1)$. At constant layer thickness, L is found to be proportional to the current I , so that $L = \beta I(e^{bd} - 1)$. The authors assume that during the formation — the current then being partly an electronic current — the collisions with the ions liberate electrons, which in turn, by excitation of the ions of foreign metals, cause the luminescence. With the formed layer, the small leakage current causes a weak luminescence. An alternating voltage of sinusoidal or rectangular shape gives a flash of luminescence at every reversal of the polarity. The intensity of this flash is found to decrease as a function of time. This is the reason why, if an alternating voltage is applied, the curve of L as a function of I or of V describes a loop. The cathode flash is explained by assuming that the layer contains traps, which are filled with electrons as soon as the Al becomes the cathode. During this process electrons recombine with the ions of foreign metals contained in the lattice. The process is of short duration. As soon as the Al becomes positive the anode flash occurs, which is considered to be the rest of the cathode flash.

R 333: P. B. Braun: The crystal structures of a new group of ferromagnetic compounds (Philips Res. Repts. **12**, 491-548, 1957, No. 6).

In this thesis (Amsterdam, December 1956) the crystal structures of four new compounds are described, the study of which has contributed in an essential way to the development of the hexagonal ferromagnetic iron compounds known as ferroxplana

(see Philips tech. Rev. **18**, 145-154, 1956/57, No. 6). The new compounds are: $W = BaFe_2^{II}Fe_{16}^{III}O_{27}$, $X = Ba_2Fe_2^{II}Fe_{28}^{III}O_{46}$, $Y = Ba_2Zn_2^{II}Fe_{12}^{III}O_{22}$ and $Z = Ba_3Co_2^{II}Fe_{24}^{III}O_{41}$; the chemical formulae of two of them were found simultaneously with the structure. The structures of the new compounds and the magnetoplumbite structure are mutually related. Oxygen and barium atoms together form a slightly expanded close-packed arrangement, with the barium atoms in certain selected positions, and the smaller ions in certain of the holes between the large ones. In all of these structures, "plates", either four or six oxygen layers thick, can be distinguished, which have very nearly the spinel structure. The relations between the structures are discussed from two different points of view. In the appendix a specially developed machine is described for the automatic summation of Fourier series with 60 cosine- and 60 sine-terms.

R 334: L. J. van der Pauw: A method of measuring specific resistivity and Hall effect of discs of arbitrary shape (Philips Res. Repts. **13**, 1-9, 1958, No. 1).

A method of measuring specific resistivity and Hall effect of flat samples of arbitrary shape is presented. The method is based upon a theorem which holds for a flat sample of arbitrary shape if the contacts are sufficiently small and located at the circumference of the sample. Furthermore, the sample must be singly connected, i.e., it should not have isolated holes.

R 335: H. G. Beljers: Determination of the gyro-magnetic ratio and the magnetic resonance damping coefficient of ferrites (Philips Res. Repts. **13**, 10-16, 1958, No. 1).

Resonance experiments are described on polycrystalline ferrite spheres in a microwave cavity with a linearly polarized magnetic field. Fundamental quantities, like the g -factor and the damping coefficient of the ferrite, are derived from the measurements. The damping coefficient can be calculated either from a line-width measurement or from the maximum amount of absorption in the ferrite at resonance. For a few samples reasonably corresponding results by the two methods are given.

R 336: P. Penning: Precipitation of nickel and copper from supersaturated solutions in germanium (Philips Res. Repts. **13**, 17-36, 1958, No. 1).

During the anneal of germanium samples supersaturated with nickel, unidentified acceptors with a low activation energy are observed, besides the acceptor levels arising from substitutional nickel atoms. Their density increases rapidly in the beginning of the anneal and reaches a value comparable to the density of nickel atoms originally present in the sample. In spite of this, the total density of acceptors ionized at room temperature remains approximately constant. Upon further annealing, the density of acceptors associated with substitutional nickel atoms and the density of the unidentified acceptors decrease exponentially with the same time constant. To explain this, a theory is put forward in which the new acceptors are assumed to be associated with vacancies left behind in the lattice when a substitutional Ni-atom jumps to an interstitial site. The interstitial Ni-atoms diffuse rapidly to nuclei of precipitation. The ratio between the densities of substitutional Ni-atoms and vacancies reaches a constant value K during the first period of anneal. According to the theory, the rate of precipitation as observed in the second part of the anneal depends on the rate of removal of vacancies, thus making it structure-sensitive, and on the value of K . A similar process is proposed for the precipitation of copper in germanium. The value of K for copper appears to be larger than for nickel. The vacancy acceptor level is found to be located at 0.02 eV from the valence band. The presence of a high density of vacancies during the precipitation of one of the two elements, copper or nickel, greatly facilitates the penetration of substitutional atoms of the other element into the sample. It is shown that concentrations much larger than the saturation concentration may be obtained. Several secondary effects of this interaction between copper and nickel are observed. The results are in good agreement with the theory. Finally it is shown how this rapid penetration of substitutional atoms in the presence of vacancies may have influenced the experimental results of other authors.

R 337: S. Duinker: General properties of frequency-converting networks (Philips Res. Repts. **13**, 37-78, 1958, No. 1).

In this thesis (Delft, June 1957; see also **R 339**, some general properties of frequency-converting networks (i.e. networks used for the purpose of modulation, mixing, detection, etc.) are analysed. These networks generally contain non-linear elements. Properties of elements satisfying the condition of "local passivity" are analysed. It is assumed

that the fundamental state, resulting from the presence of energy sources, is subjected to a small disturbance. The first-order perturbational equations are linear differential equations with time-dependent coefficients which satisfy the reciprocity condition. These equations are further analysed, assuming a periodic fundamental and perturbational state and after the introduction of complex quantities. The network equations can then be represented with the aid of infinite matrices. The effect on the matrix equations is investigated of the shifting of the time origin, of the interchanging of the role of the signal frequency and a frequency generated by conversion, of a symmetric fundamentals state and of the presence of linear impedances. After introducing the notions of "fictitious" meshes and ports it is shown that the equivalent circuit of a conversion network includes network elements that are not physically realizable and even may be active. An energy theorem for purely-reactive conversion networks is derived and the possibility of instabilities, the conversion gain, and the condition for the absence of reaction on the signal source are discussed. The simplification of the matrix equations resulting from a balanced configuration of elements is discussed with the aid of some examples.

R 338: P. Penning: Generation of imperfections in germanium crystals by thermal strain (Philips Res. Repts. **13**, 79-97, 1958, No. 1).

Thermal strain is induced in a crystal if it is heated inhomogeneously. These strains can be so large that either plastic flow or cracking may take place. Both effects have been observed in crystals quenched from high temperature. In the case of plastic flow the distribution of dislocations, as revealed by etching, shows marked characteristics, dependent on the manner of quenching and the orientation of the crystal. It is shown that these characteristics can be explained very well if it is assumed that the amount of plastic flow is directly proportional to the elastic strain induced by the non-uniform temperature. During the growth of a crystal from a melt, thermal strain may also very well be present in the crystal. The main sources of these strains are indicated and their influence on the perfection of the growing crystal are discussed. Secondary effects of plastic flow due to thermal strain in grown crystals and quenched samples, such as residual stresses and the freezing-in of defects, are discussed briefly. See also Philips tech. Rev. **19**, 357-364, 1957/58 (No. 12).
